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FATE OF RADIOACTIVE CONTAMINANTS IN
WATER

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Robert A. Taft Sanitary Engineering Center

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FATE OF RADIOACTIVE CONTAMINANTS IN WATER

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Radiological Health Research Activities

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE

Public Health Service
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Division of Radiological Health

Robert A. Taft Sanitary Engineering Center
Cincinnati 26, Ohio

1959

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SUMMARY

This report on water borne radionuclides records progress in four specific areas of study during the first year of the project--instrumentation, tritium, estuarine and marine waters and biota, and the fate of radioactive material.

The first section describes specialized instrumentation provided to carry out the studies for the Atomic Energy Commission, and the facilities available for housing the instrumentation. The instrumentation includes a 100-channel pulse height gamma spectrometer, low background beta anti-coincidence counter, and liquid scintillation spectrometer. A six-inch steel shield, having interior dimensions 18" x 18" x 24", houses the 4" diameter x 4" thick solid sodium iodide (thallium activated) gamma scintillation crystal and 5" photomultiplier tube.

Because conventional methods for gross measurement of alpha or beta-gamma activity are not suitable for the detection of tritium, contained in the wastes from PWR, Shippingport, a project was initiated to develop methods for its identification. Liquid scintillation counting was selected because levels at approximately 1/1000 of the MPC value ($2 \times 10^{-4} \mu\text{C}/\text{ml}$) could be measured. Two scintillation solvents were chosen: the first contained dioxane, PPO, POPOP*, and naphthalene; the second contained dioxane, anisole, dimethoxyethane, PPO, and POPOP. Tritium levels found in a single series of four samples from Shippingport are reported.

As a result of nuclear powered vessel construction, information is desired on methodology for identifying specific radionuclides in estuarine and marine waters and biota. Furthermore, the utilization of the oceans for waste disposal makes mandatory suitable methods for measuring radioactivity to control such operations. This section of the report presents a review of current practice in this field and a description of our approach toward elucidating the problem.

The final section gives the results of analyses on a single sample of effluent from KAPL and describes the techniques to be employed in determining the fate of specific radionuclides in the stream environment.

* PPO is 2,5 diphenyloxazole; POPOP is 1,4-di [2-(5-phenyloxazolyl)]benzene.

I. INSTRUMENTATION

Byron M. Branson and Gerald J. Karches, Physicists

1. INTRODUCTION

In order to carry out the work under contract from the AEC, certain specialized instrumentation was deemed necessary. This instrumentation included the 100-channel gamma spectrometer, equipped to handle large samples in a six-inch steel shield; a low-background steel-shielded anti-coincidence beta counter; and a liquid scintillation spectrometer suitable for counting tritium.

2. COUNTING ROOM

Because these instruments were to be used at extremely low levels, it was necessary to install them in a special low level counting room. To accommodate instruments provided with heavy external shielding, this room had to be at ground level and specially constructed.

The room consists of an area approximately 10-1 2' x 24' in the third basement of the Sanitary Engineering Center. One wall of this area is the external wall of the building basement, of heavy poured concrete construction, in contact with earth throughout its height and length. Two new walls were provided of 8" concrete block construction, the cores filled with concrete to a height of 10 feet; the fourth wall is an existing 8" poured concrete wall. The ceiling is of reinforced concrete 12" in thickness. The door area is the only substantial "hole" in the room shielding. The instruments have been grouped in the more heavily shielded area away from the door.

The utilities provided are electricity, air conditioning, water, and drainage. The electrical supply consists of three separate 110-volt lines with a total capacity of 90 amperes. Each instrument is equipped with a Sola transformer to maintain constant voltage, and some have filter chokes to prevent spurious counts from line transients. The room is illuminated by incandescent flood-light-type bulbs, since it has been reported that fluorescent light induces

a photo-luminescence in glass which would interfere with the counting of samples in the liquid scintillation counter. A three-ton water-cooled air conditioner was installed to dissipate the heat produced by the electrical instrumentation. To prevent accumulation of stagnant air, a small exhaust fan has been located high in the wall where it will contribute to the maintenance of suitable temperature and humidity conditions. The water and drainage facilities are primarily for the air conditioner, but a small sink is provided for washing sample bottles and the like. The floor is covered with asphalt tile, and the ceiling with acoustical tile to minimize air conditioner noise.

3. 100-CHANNEL GAMMA SPECTROMETER

Many of the radionuclide analyses will be facilitated by the use of a 100-channel gamma spectrometer to identify the various gamma-emitting nuclides present and in some cases to permit their direct quantification. After considerable investigation it was decided that a 100-channel spectrometer of the Argonne National Laboratory type would be most suitable, and such a unit was obtained commercially.* This unit (shown in Fig. 1) consists of a precision high voltage supply, a linear amplifier, a pulse converter, and a magnetic core address and memory unit, together with associated power supply and read-out equipment. Three types of read-out are available--an oscilloscope presentation, a digital print-out, and a spectrum plotter. The first two of these have been obtained. A camera to photograph the oscilloscope record has also been purchased.

To date, spectra have been obtained essentially with 2" diameter by 2" thick sodium iodide (thallium activated) crystals, either solid or well type. Some of these spectra are shown in Figures 13 and 14.

A 4" diameter x 4" thick solid scintillation crystal will be used with a 5" photomultiplier tube. This detector unit (Figure 2) is installed

* Radiation Instrument Development Laboratories.

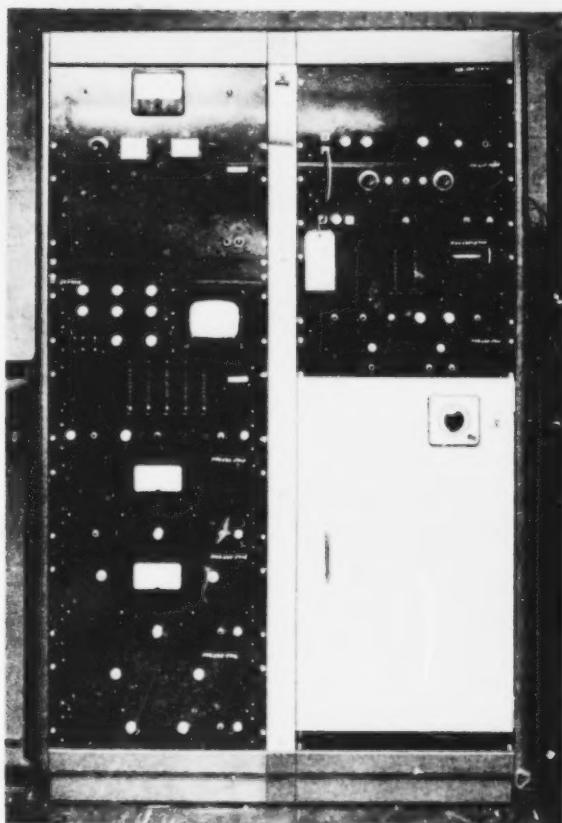


Figure 1. 100-CHANNEL GAMMA SPECTROMETER

on shelves fastened to the hinged door of the 6" steel shield (Figure 3), which was provided to reduce background. The shield has interior dimensions of 18" x 18" x 24". Before assembly, the steel plate used in the construction of the shield was found to be contaminated to a significant extent with zirconium and ruthenium, presumably from fallout. Sand-blasting adequately removed the contamination, as indicated by the background spectrum.

Thus far, the gamma spectrometer has been used chiefly for the identification of radionuclides. Quantitative determinations will depend on adequate calibration of the equipment. The response of the instrument has been determined to cesium-137, cerium-144, zirconium-niobium-95, ruthenium-rhodium-106, barium-lanthanum-140, and various mixtures of these. Quantitative determinations require absolute calibration of the solutions either with a 4-pi counter or with purchased calibrated solutions. A 4-pi counter was obtained in February; cer-

tain difficulties relating to charge build-up and sample preparation have been resolved, and the instrument is available for use.

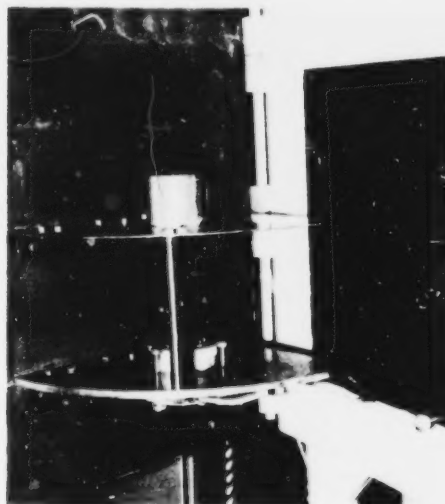


Figure 2.
INTERIOR OF SHIELD SHOWING DE-
TECTOR ASSEMBLY ATTACHED TO
HINGED DOOR OF 6" STEEL SHIELD.

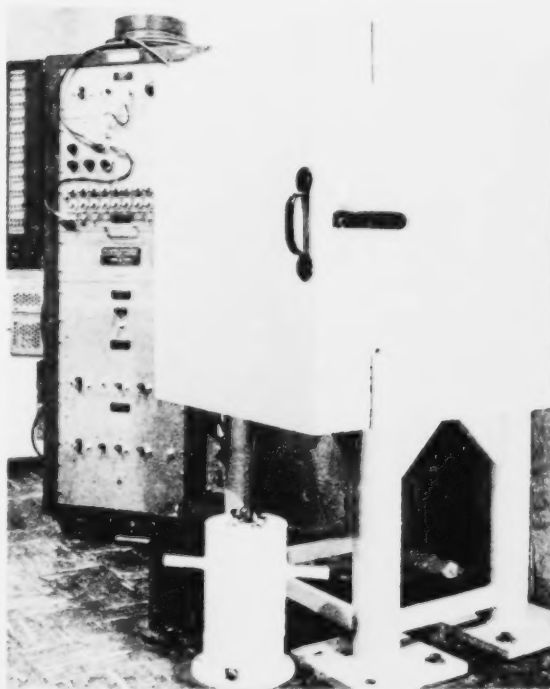


Figure 3. EXTERIOR OF SHIELD FOR
USE WITH GAMMA SPEC-
TROMETERS.

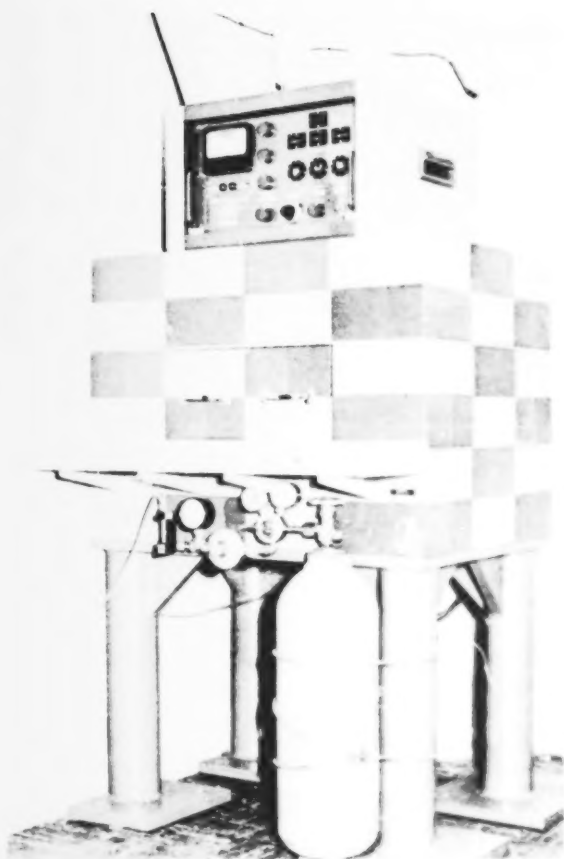


Figure 4. LOW BACKGROUND ANTICOINCIDENCE BETA COUNTER.

4. LOW BACKGROUND BETA COUNTER (Figures 4 and 5)

Use of the low background beta counter has been delayed by construction in the low level counting room. Preliminary work has established that the instrument plateau and background are satisfactory; in fact, the background in each chamber is less than half the specified 1 cpm. Evaluation of the instrument will proceed through the use of the 4- π counter and the commercially available solutions.

5. LIQUID SCINTILLATION SPECTROMETER (Figures 6 and 7)

A liquid scintillation spectrometer was selected for the determination of tritium. The



Figure 5.
LOW BACKGROUND ANTICOINCIDENCE BETA COUNTER. INTERIOR VIEW. Rolling doors are opened to show counting chambers. Chamber A on left is opened and shows one anticoincidence tube of the ring which forms an umbrella around the end window G-M tubes. Chamber B on the right is closed.

reasons for this choice are given in the section on tritium (Section II of this report). The instrument was put into operation in February, but its use has been restricted by construction work. Beforehand, a physicist and chemist had been assigned to Professor C. G. Bell at Northwestern University to gain experience in operation of the equipment, with particular reference to tritium analysis. This training permitted more rapid utilization of the liquid scintillation spectrometer.

In spite of construction delays, operating characteristics of the liquid scintillation spectrometer have been determined. A number of isotopes, including tritium, have been counted under varying voltage and discriminator settings with two solvents, one consisting

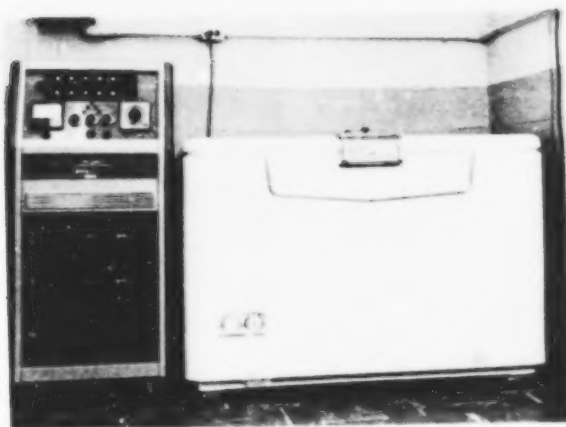


Figure 6.
LIQUID SCINTILLATION SPECTROMETER. Beta counter for tritium and carbon-14 determinations. Exterior view of freezer and control panel with scaler at left.

essentially of water in dioxane and the other of water in a mixture of dioxane, anisole, and dimethoxyethane ("611").

6. SUMMARY

Necessarily, this period has been devoted largely to initial tooling up on instrumentation. Valuable experience has been gained in the operation and maintenance of the highly spe-

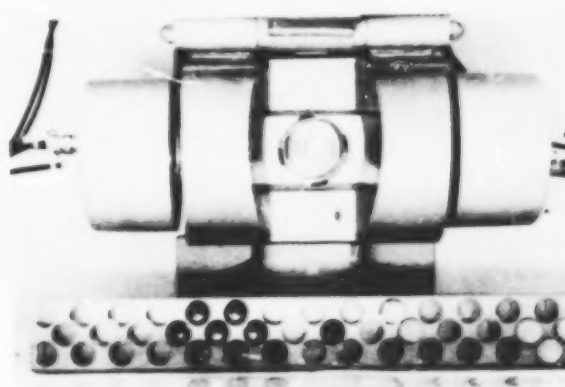


Figure 7.
LIQUID SCINTILLATION SPECTROMETER. INTERIOR VIEW. Sample chamber is shown in the center, and the tubes are at each end of the assembly. The sample container is housed in a 2" lead shield.

cialized equipment, enabling its more efficient and more accurate use.

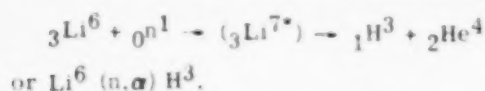
Additional calibration and evaluation studies are necessary and will be made while the instrumentation is in use for research and routine measurements. Better methods of sample preparation, mounting, and counting are desirable and will be studied.

II. TRITIUM

Seymour Gold and Vincent J. Sodd, Chemists

1. INTRODUCTION

It has been reported that tritium will be discharged in the primary coolant from the PWR at Shippingport, Pennsylvania. This isotope arises from neutron irradiation of lithium introduced in conditioning the coolant to inhibit corrosion. Tritium is produced by an (n, α) reaction:



The concentration of tritium in the reactor coolant depends on the product of power level and length of exposure.

Since tritium has a half-life of more than 12 years, very little decay will take place in any reasonable holdup time. Being a constituent of water, it cannot be removed by distillation, chemical treatment, or ion exchange. Fortunately, tritium is eliminated from the body fairly rapidly. This fact, together with the extremely low energy of its beta radiation, accounts for its high maximum permissible concentration, 0.2 μC ml.

In the Industrial Waste Permit for PWR, Shippingport, the State of Pennsylvania agreed to a daily discharge of 10 curies of tritium over any 365 consecutive days, with a maximum tritium discharge of 50 curies in any one day.

On the basis of discharge of 10 curies of tritium per day to the Ohio River and an assumed average flow of 25,000 ft³ sec, there would be a tritium concentration of 1.6×10^{-7} μC ml or approximately 0.3 d m ml, which is approximately 0.0001 per cent of the MPC value. Detection at this level would be impossible without prior concentration of the tritium. However, in the condenser cooling water, for a flow of 58,000 gpm, 10 curies day would give a concentration of 3×10^{-5} μC ml or 60 d m ml which may be detectable.

The project has a two-fold purpose: (1) to develop and evaluate methods for the routine

analysis of tritium in water at levels well below the MPC; and (2) to study the effect of tritium on assay procedures presently used in radiological analyses.

In the subsections that follow, a review is presented of the techniques used for the determination and measurement of tritium and the results that have been obtained to date.

2. PRIOR WORK

Following numerous searches for a stable isotope of hydrogen of atomic weight 3, Alvarez and Cornog in 1939 discovered tritium and noted its radioactive nature. Tritium was later found in extremely small quantities in nature as a result of cosmic ray bombardment of the atmosphere. Investigation of the radioactive characteristics of tritium showed that it was a pure beta emitter of extremely low energy, the maximum energy being only about 18 kev. Its half-life of approximately 12.5 years was established by Jenks, et al.

Because of their extremely low energy, tritium beta particles cannot be counted in conventional equipment. The range of an 18 kev beta particle is only about 0.6 milligrams per square centimeter (in aluminum). Even thin windows, which usually run about 1 to 2 mg sq cm, cut out tritium radiations, while in a windowless counter there is extremely low counting efficiency for tritium in conventionally prepared samples because of self-absorption, the radiations in this case being yet more strongly absorbed than alpha particles. For these reasons the first interest in counting tritium, except at extremely high activities, was limited to gas-phase counting.

Internal sample, gas-phase counters are quite satisfactory for the determination of tritium. The beta particles originating in the gas volume are absorbed in the gas, leaving an ionization track which can be collected and measured by conventional ion chamber, proportional chamber, or Geiger techniques. In practice, there are certain difficulties with the

technique. In the first place, the gaseous sample having low density is normally quite small. Even at atmospheric pressure, a liter of gas contains less than one gram of water or the corresponding amount of hydrogen. Furthermore, water vapor is a very unsatisfactory sample for counting in either the Geiger or proportional regions. For this reason, water samples containing low levels of tritium often have been converted to gaseous hydrogen, which is counted in a large chamber operated in the Geiger region. In this manner Libby and his co-workers and Brown and Grummitt in Canada analyzed the tritium content of rain and other natural waters.* This method required a cumbersome chemical conversion to hydrogen gas and the use of large counters which would normally have a very high background. For the determination of low levels of tritium it was necessary to reduce background by very rigid shielding and anti-coincidence techniques. This is, however, the most sensitive technique for the assay of tritium at very low levels.

In the late 1940's the availability of better photo-multiplier tubes and the discovery that certain materials such as anthracene and thallium-activated sodium iodide would fluoresce under ionizing radiation led to revived interest in scintillation techniques. It was discovered somewhat later that solutions of various organic materials would scintillate under the influence of alpha, beta, and gamma radiation, and that these scintillations could be observed by photomultiplier tubes. This development was of extreme interest in the counting of tritium and other weak beta emitters. It offered the possibility of obtaining the advantages of gas counting, i.e., 4- π geometry and no self-absorption losses, together with the advantages inherent in handling a liquid rather than a gaseous phase.

The earliest liquid scintillation solutions employed solvents such as toluene and xylene, which by themselves were unsuitable for use with water or aqueous media. Efforts to dissolve water in these liquids were successful with the addition of mutual solvents such as ethyl alcohol and dioxane. Still later it was found that solutions could be based on dioxane instead. However, the quantity of water that could be dissolved was limited by (1) the possibility of precipitating out some of the scintillating solutes which are water-insoluble complex

organic molecules and (2) the fact that the size of the light pulses produced is severely reduced by foreign materials, including water. Concentrations of water therefore have been limited to not more than 10 to 20 per cent, depending on the specific scintillating mixture used.

3. CALIBRATION

On the basis of the prior work reported, it appeared that for the determination of tritium only two methods should be seriously considered, liquid scintillation and gas phase counting, preferably in an internal Geiger tube. The gas phase method is somewhat more sensitive; liquid scintillation is much simpler in its sample preparation requirements. Even with the less sensitive liquid scintillation technique, it appeared that tritium could be determined at levels of approximately 1 1000 of the MPC, that is, 2×10^{-4} μ c ml or approximately 440 d m ml. There was also some prospect of increasing the sensitivity to measure the concentration expected in PWR condenser cooling water (60 d m ml), either by a somewhat more complicated sample preparation technique or through preliminary fractional distillation to concentrate the tritium. Accordingly, liquid scintillation was chosen for tritium determination.

Two scintillating solutions were chosen as the most promising for our purposes. The first of these is based on dioxane as solvent with PPO (three grams per kg) as primary scintillating solute and POPOP (0.05 grams per kg) as secondary solute along with naphthalene (50 grams per kg) to enhance the efficiency. This system is commonly used with a ten per cent water sample, i.e., 9 parts of the organic solution to one part of water by volume. The other solution is also based on dioxane as a scintillating solvent but with additional solvents to permit the introduction of greater quantities of water. The solvents are six parts of dioxane, one part of anisole, and one part of 1,2-dimethoxyethane, by volume. The scintillating solutes are again PPO (15 grams per liter) and POPOP (0.62 grams per liter). This system does not contain naphthalene. This so-called "611" solvent can be used with as much as 20 per cent water by volume, i.e., one volume of water to four volumes of organic solution. The efficiency of these solvents was determined through the use of tritiated water of known activity.

* For these samples preliminary concentration of tritium by electrolysis was required.

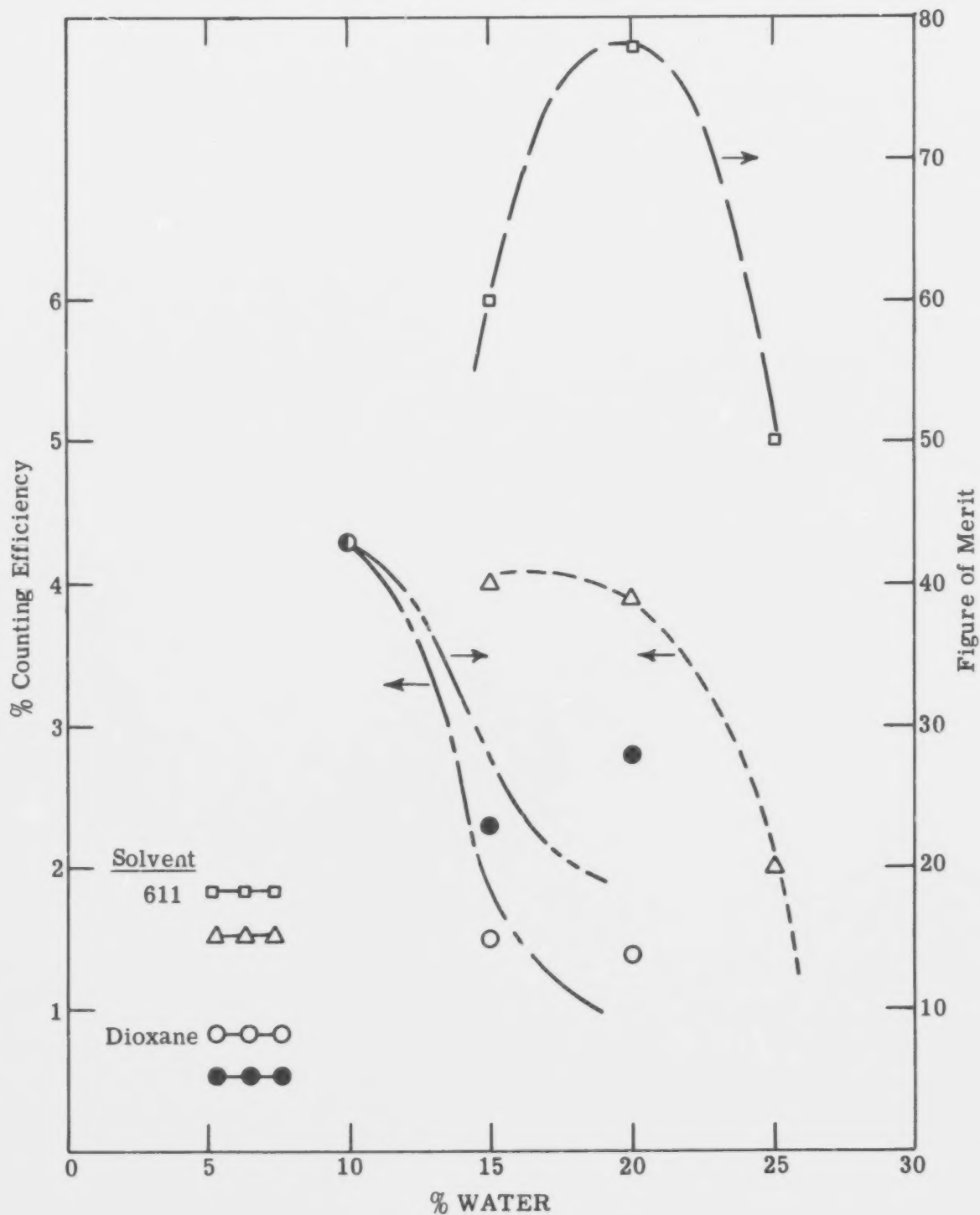


Figure 8. RELATION BETWEEN PER CENT WATER IN SCINTILLATION SOLVENT AND COUNTING EFFICIENCY OR FIGURE OF MERIT.

Variables involved in the determination of counting efficiency and overall value of a liquid scintillation counter include sample size, high voltage setting, channel width, and background. At low levels small sample size is undesirable, since the small volume of low activity water will contain only a few disintegrations per minute and this will make the detection difficult if not impossible. However, increasing the volume of water causes a decrease in the efficiency with which the disintegrations are detected. Thus, with very small samples few counts are obtained because there are few disintegrations, whereas with very large samples few counts are obtained because the efficiency falls practically to zero, and an intermediate optimum sample volume will give the greatest counting rate for a given water activity. This can be understood more clearly from the accompanying graph, Figure 8, which gives curves for the water-dioxane mixture and the "611" solvent. Two curves are given for each solution, one showing counting efficiency against per cent water, and the other the product of counting efficiency and water volume against per cent water.

Even with a fixed sample of definite activity, the counting rate can be varied by changing the high voltage to the tube and also by changing the discriminator which determines what portion of the spectrum is accepted and recorded on the scalars. At low applied voltages with low amplification, the extremely small pulses produced by the photomultiplier tube will not be large enough to be accepted by the circuitry. As the voltage is increased, the pulses will be amplified more and more until a point is reached where most of the pulses are large enough to be accepted by the discriminator. Since the discriminator is normally operated with both high and low energy cutoffs, at higher voltages many of the pulses are amplified to the point where they are too large to be accepted by the discriminator. Thus, there will be relatively few counts at low voltage settings, a higher number of counts at intermediate settings, and again a low number of counts at very high settings of the voltage. The discriminator must be operated with both a low and a high energy cutoff because the low energy cutoff eliminates much of the electronic noise in the system, which is a large part of the background, and a high energy cutoff eliminates high-energy noise contributing to the background and particularly discriminates against other higher energy emitters which may be present in the

sample. Preliminary indications are that tritium can be counted in the presence of moderate quantities of higher energy emitters such as Sr^{89} and Sr^{90} without serious interference, but data on this point are still incomplete. A number of curves of counting rate as a function of high voltage setting and of discriminator setting are presented, Figures 9-12, to illustrate these variable influences. For any discriminator setting, there is an optimum voltage setting which will give the maximum counting efficiency and therefore the maximum counting rate, for any given sample.

The changes in instrument setting which affect the counting efficiency also affect the efficiency for counting background events. The optimum settings found for instrument counting efficiencies with 20 per cent water in the "611" solvent are high voltage at Tap 9 (1280 volts) and discriminator setting of 10-100 volts. This has given a counting efficiency of 3.9 per cent with 20 per cent of water by volume, as shown in Figure 8. The 3.9 and the 20 may be multiplied to give a "figure of merit" of 78. At relatively high counting rates, the error due to the background is insignificant and the "figure of merit" is the criterion for optimum performance. At low counting rates, however, the error due to background becomes significant and the figure of merit calculated above must be corrected by dividing the square root of the background rate. Therefore it was important to determine the variation of background with instrument settings and with water content of the solutions. Neither influence was found sufficient to change the conclusions reached from the preceding work--that the most suitable tritium determination conditions were (1) water 20 per cent of solution by volume, (2) "611" solvent, (3) high voltage tap 9, and (4) window discriminator setting 10-100 volts. Under these conditions and with the assumption of a background of 50 cpm measured over 1000 minutes, the calculated minimum detectable tritium activity in water varied from $9.2 \mu\mu\text{c/ml}$ to $1.8 \mu\mu\text{c/ml}$, depending on the counting time over a range of 20 to 1000 min. Since the MPC for tritium, from Handbook 52, is $0.2 \mu\text{c/ml}$ or $200,000 \mu\mu\text{c/ml}$ this would enable detection of as little as 0.0009 per cent to 0.0046 per cent of the MPC. These detectable activities are in the range to be expected in condenser coolant if ten curies are discharged daily.

It has recently been found that the counting efficiencies of these solutions as originally

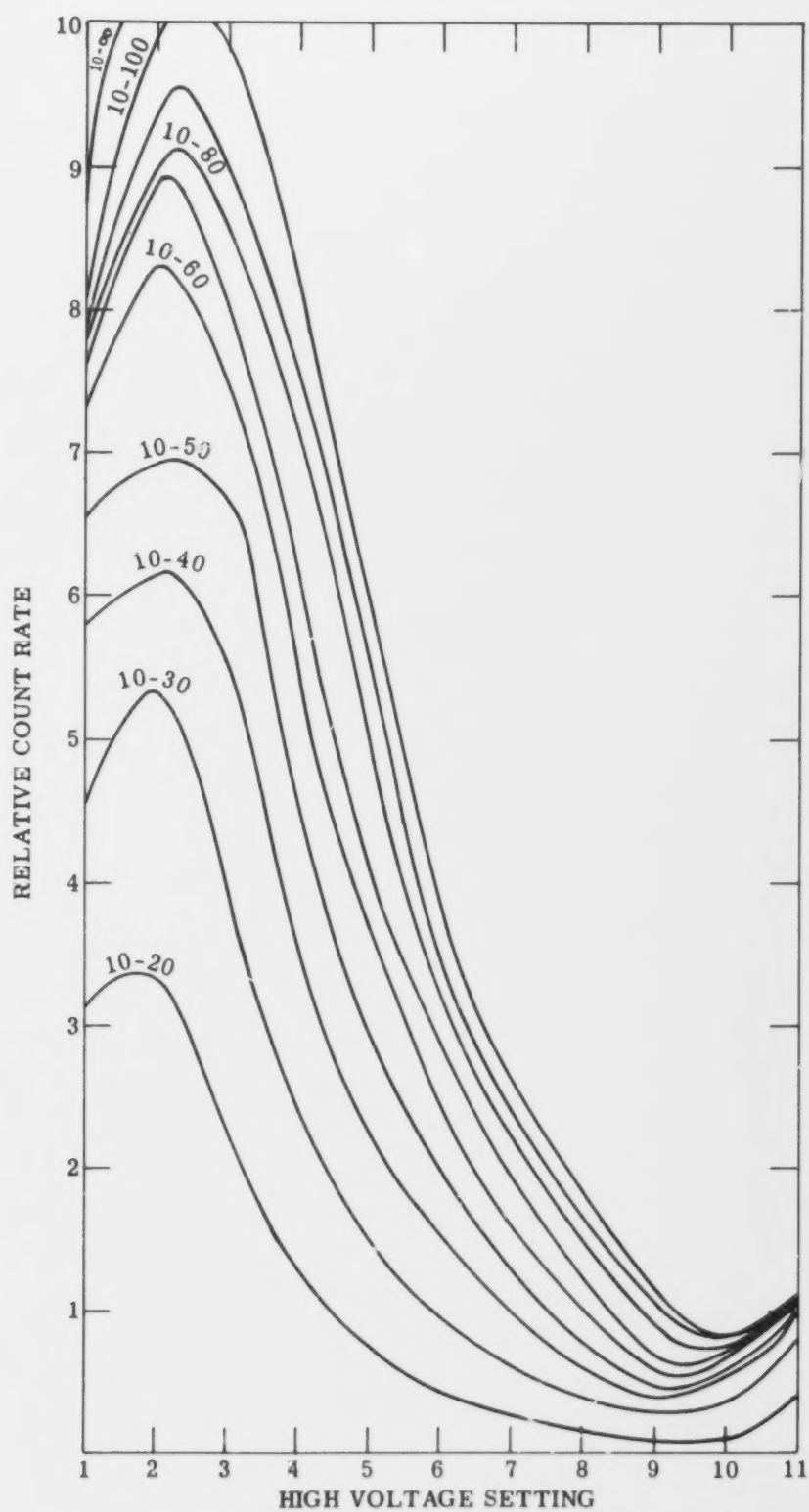


Figure 9. TOTAL COUNTS VS HIGH VOLTAGE SETTING - STRONTIUM-89.
Solvent: Dioxane - iter 10%

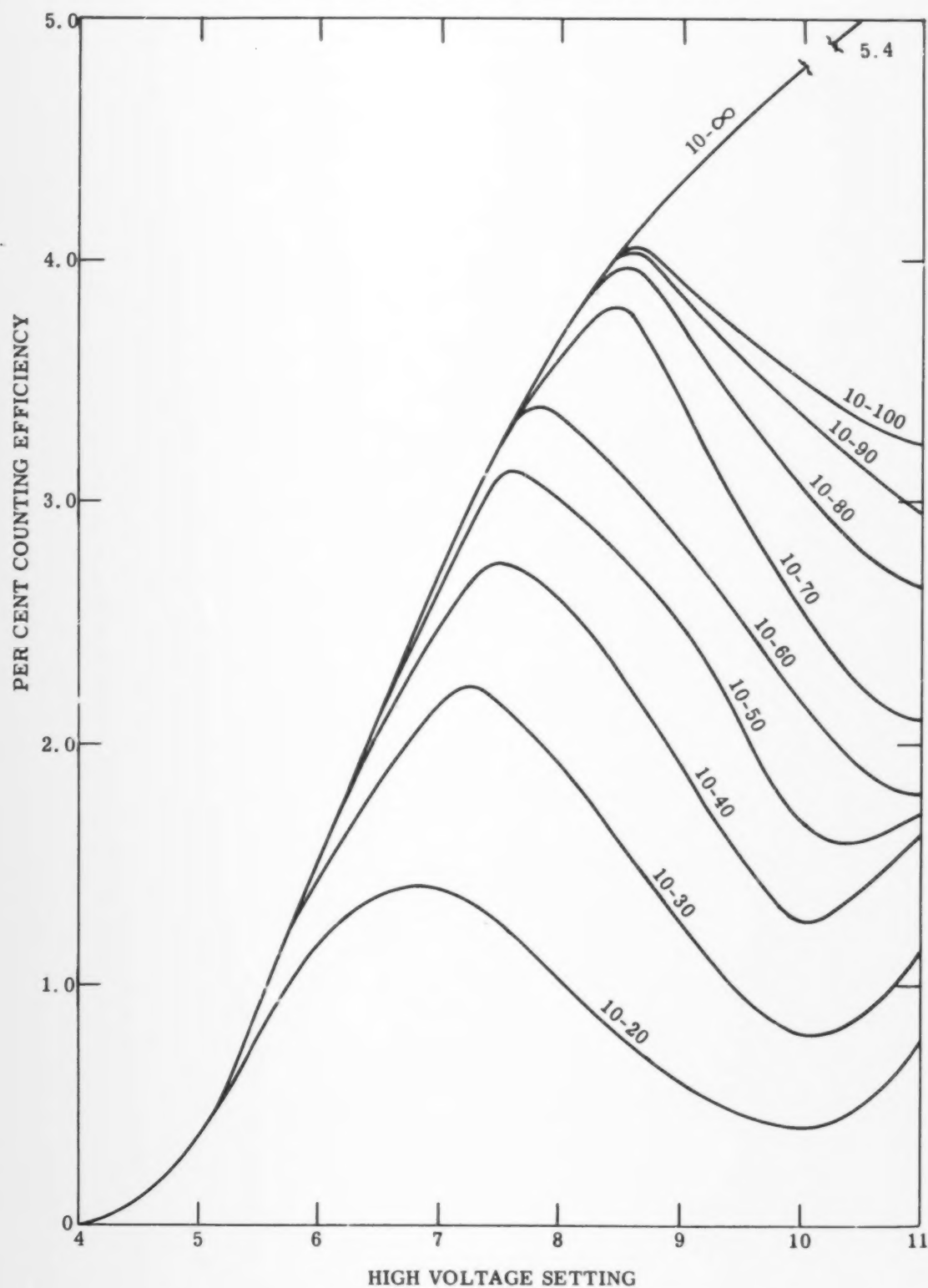


Figure 10. COUNTING EFFICIENCY VS HIGH VOLTAGE SETTING - TRITIUM
"611" Solvent

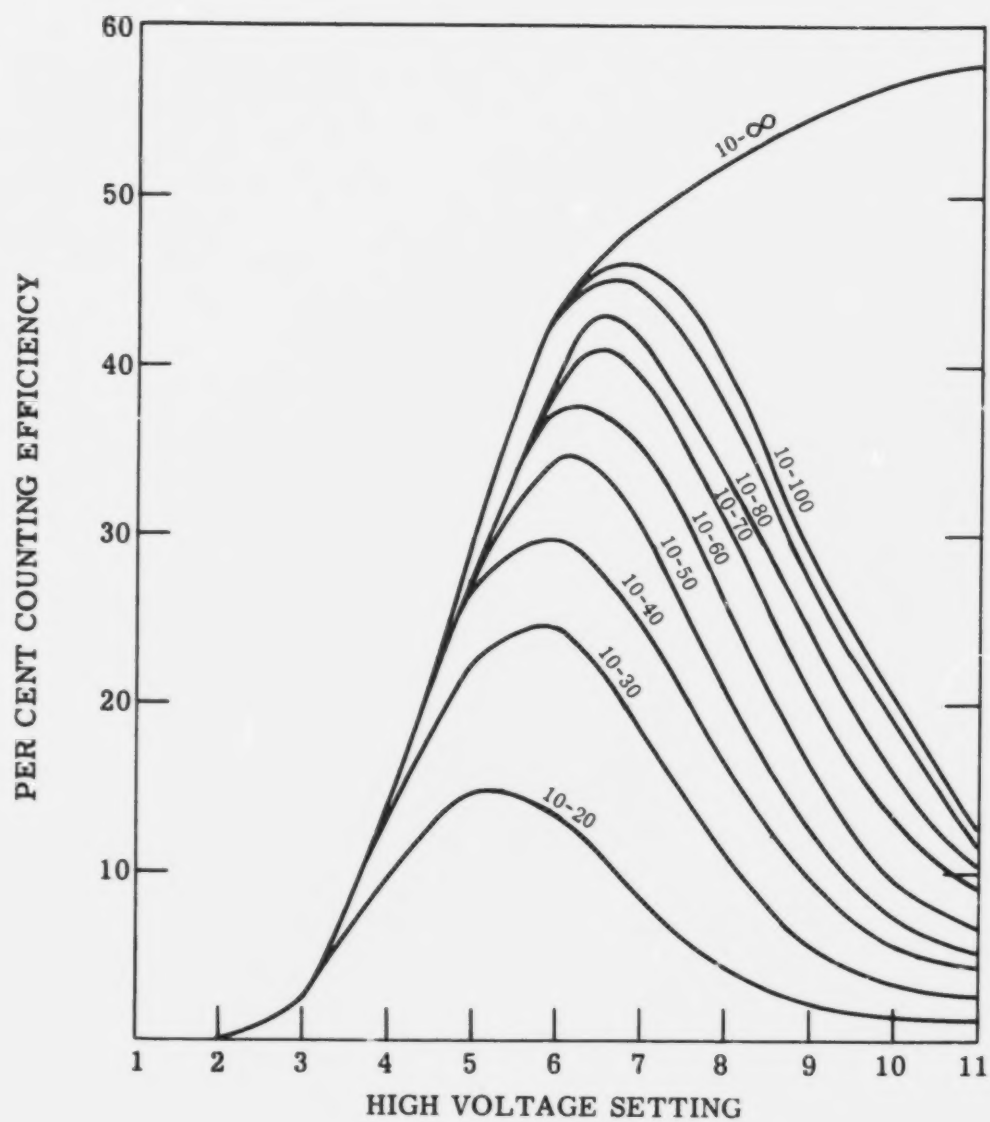


Figure 11. COUNTING EFFICIENCY VS HIGH VOLTAGE SETTING - CARBON-14
"6:1" Solvent

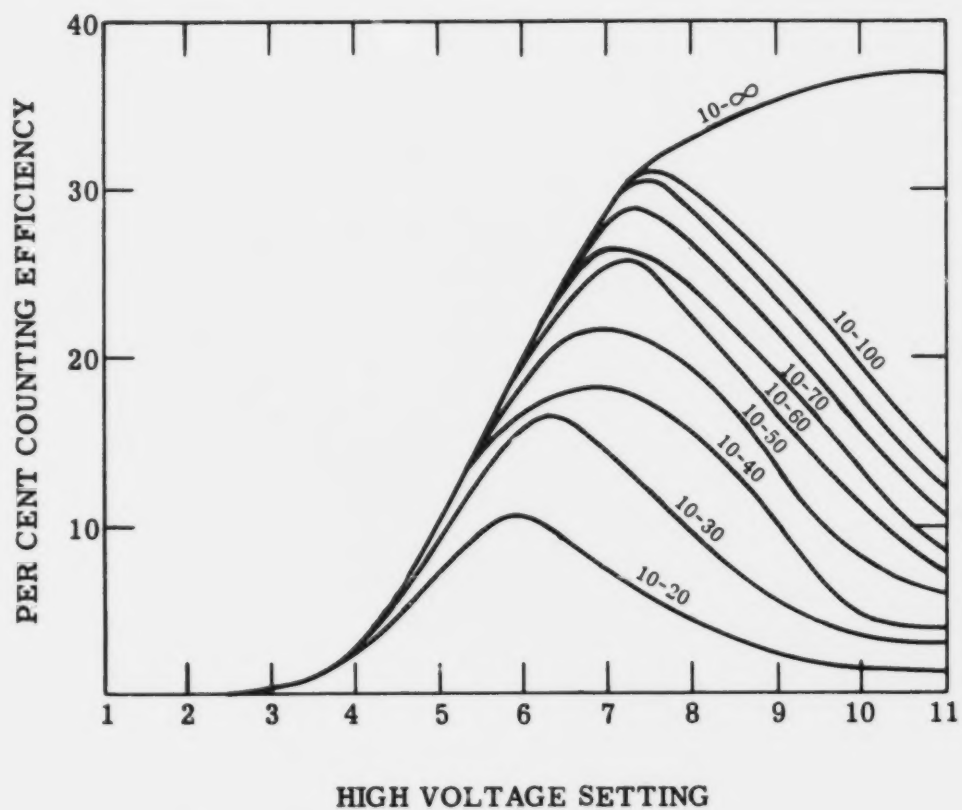


Figure 12. COUNTING EFFICIENCY VS HIGH VOLTAGE SETTING - CARBON-14
Dioxane - Water (20% Water)

measured are no longer maintained, but decrease as the organic solution stands. With this lowered efficiency the minimum detectable activity rose to about 2.7 to 14.0 $\mu\text{C/ml}$, which is still only about 0.002 to 0.007 per cent of the MPC. The cause of the drop in counting efficiency of the solutions is not known, but it may reflect the extremely rigorous purity standards generally required in scintillation work. The solvents procured as the best commercially obtainable were not further purified. It is hoped that by rigorous purification of these solvents the high efficiencies originally obtained will be maintained over a longer time.

Values of counting efficiency as a function of solvents used are summarized in Table 1, and the effect of fluorescent light on background counting is presented in Table 2.

4. MEASUREMENTS

To date, one set of samples has been received from Shippingport for analysis. This consisted of four samples of approximately two liters each, representing primary coolant, condenser coolant, river upstream, and river downstream, collected on March 10 and 11, 1958. Tritium was determined by counting a four-ml sample together with 16 ml of "611" scintillation solution in the liquid scintillation spectrometer. To determine the efficiency of the instrument,

known samples were prepared and counted concurrently. Tritium was detected only in the primary coolant, the amount found being 29.3 $\mu\text{C/l}$, approximately 1.7 of the MPC. The sample, when received, was marked as having 25 $\mu\text{C/l}$; however, it was not known whether this was intended to represent an accurate analysis. In addition to the determination of tritium, for completeness, gross beta analyses of all the samples were made by evaporating a volume of 250 or 500 ml in the conventional SEC manner. The low value obtained for primary coolant ($< 2.5 \times 10^{-5}$ of the tritium activity) indicates that tritium in this low-solids sample does not interfere with the gross non-volatile beta assay. When the gamma spectrum of the primary coolant sample was measured, no definite gamma activities could be identified. The results of these analyses are summarized in Table 3.

5. FURTHER WORK

Besides a stepped-up schedule of analyses, the tritium program must be further developed in several directions. First, the causes of the variation of counting efficiency with time must be determined, and such variation prevented. Possibly this research will also yield higher counting efficiencies. Second, work is needed to extend the sensitivity of the method to levels little higher, if any, than the natural tritium background in ordinary water. One possibility

Table 1. EFFICIENCY OF TRITIUM COUNTING

Sample No.	Description	Counting efficiency ^a , %		
		March 11	April 15	April 27
Tc-124-611	"611" Std.	4.02	3.36	--
Tc-151-611	New "611" Std.	--	--	2.57
Tc-124-B10	D-W ^b Std.	4.46	1.69	--
Tc-151-B	New D-W Std (10%).	--	--	1.86
		Counts per minute		
D-W(20%)	Background.	46.0 $\pm$ 0.5	40.2 $\pm$ 5.7	
D-W(10%)	Background.	38.0	37.6	

a At high voltage tap 9, Channel 10-100.

b Dioxane-water.

Errors are counting errors only, at 95% level.

FATE OF RADIOACTIVE CONTAMINANTS IN WATER

Table 2. EFFECT OF SAMPLE EXPOSURE TO FLUORESCENT LIGHT

Time after exposure ^a	Length of count, min.	Background counting rate ^b , cpm
Before exposure	1000	50.1 ± 0.4
Immediately	10	170 ± 8.1
1 hour	1000	52.1 ± 0.4
23 hours	150	51.7 ± 1.1
25 hours	1000	50.9 ± 0.4

a At high voltage tap 9, Channel 10-100.

b To start of counting time. Errors are counting errors only, at 95% level.

here is the use of larger samples, since 50 or possibly 75 ml instead of the present 20 ml can be counted in the existing apparatus, but efficiency with the larger samples is somewhat lower and background is somewhat higher. Another possibility is conversion of the water to an organic liquid more easily adapted to the conventional toluene-based scintillating solu-

tions. Some work has been done in Israel on the exchange of tritium from water with the hydrogen of toluene. Since the toluene can then be used as the scintillation solvent, larger samples can be handled with no loss of efficiency. This method is contemplated for use with only a few special samples, since preparation time is of the order of 100 hours.

Table 3. ANALYSES OF SHIPPINGPORT SAMPLES

Sampling point	Date collected	Tritium $\mu\text{C l}$	Gross beta ^a $\mu\text{C l}$
Primary Coolant ^b	3 11 58	29.3 ± 0.15	680 ± 23
Condenser Coolant.	3 11 58	< 0.016	5 ± 7
Montgomery Dam	3 10 58	< 0.013	4 ± 7
Lock 7	3 10 58	< 0.013	3 ± 6

a Non-volatile beta only.

b "B" sample train. Errors are counting errors only, 95% level.

III. ESTUARINE AND MARINE WATERS AND BIOTA

Vincent J. Sodd and Richard J. Velten, Chemists

1. INTRODUCTION

As submitted, this project was intended to (1) develop and evaluate methods for the determination of gross radioactivity in water of high (>1 per cent) salt content; (2) develop methods for identification and determination of specific radionuclides in such water; (3) develop methods for radioactivity determination, both gross and isotopic, in estuarine biota; and (4) study the possibility of utilization of off-shore waters for disposal of radioactive wastes.

Interest in the effect of radioactive materials in estuarine and marine environments has been stimulated by the rapid strides being made in the utilization of nuclear energy for the propulsion of ships, by the probability that radioactive waste discharged from atomic power stations into streams will eventually reach the sea through estuaries, and by the introduction of radioactive materials into saline waters by weapons tests and waste disposal operations.

The determination of radioactivity in saline waters is enormously complicated by the high solids content. Simple evaporation methods developed for fresh waters are not applicable to saline waters. The radionuclides of interest must be separated from the major ionic components of salt water: sodium, magnesium, calcium, potassium, chloride, sulfate, and bicarbonate.

The uptake of various radionuclides by plankton and other biota in estuarine and marine environments is an important factor in evaluating the effects of radioactive contamination of these environments. Biological concentration of radionuclides may necessitate more stringent control of radioactive materials, particularly where food organisms are involved. This approach may also permit more sensitive determination of contamination through the testing of indicator organisms. Conversely, nuclides which do not enter the biological chain may have much less impact on the inhabitants of the environment.

Since the levels of activity in many of our estuarine waters are assumed to be very low, preliminary studies will be slanted toward determination of the stable elements concentrated by marine biota. Application of sophisticated instrumentation, including gamma spectrometers and low-background beta counters, to reduce sample preparation problems, will be a major part of these developmental studies.

In studying the possible use of off-shore waters for disposal of radioactive waste, the initial phase will include a survey to identify and classify off-shore water uses. These data will be obtained from other agencies through the PHS Regional Offices.

2. PRIOR WORK

The determination of radioactivity in salt water and in marine organisms has been performed in connection with nuclear weapons detonations. Besides the many measurements sponsored by the AEC, this includes considerable investigation by Japanese workers, particularly as applied to organisms for human food.

Several methods have been utilized for determination of radioactivity in sea water. Although the earlier, simpler methods were less sensitive than the later, more complicated methods, they were suitable for water of rather high radioactivity content. Thus, in the early surveys by the University of Washington group, 5-ml and later 10-ml portions of sea water were evaporated. In late 1955, 100-ml samples were tested by treatment with sodium carbonate and measurement of the radioactivity in the resulting precipitate. Approximately 77 per cent of mixed fission product tracer was recovered by this technique. In a modification of this procedure, 250-ml samples were tested by adjusting the pH so as to precipitate only calcium when carbonate was added, the other cations remaining in solution. The Japanese have precipitated radioactivity from liter samples of sea water by the addition of carrier iron and barium followed by adjustment of the pH to a phenolphthalein endpoint. They reported this collected 80 per cent or more of the total activity.

Where radiochemical analysis of the water rather than a gross activity count is desired, a scheme used by the Japanese may be mentioned. They added iron and lanthanum to a liter of sea water, precipitating these as hydroxides. An alkaline earth fraction was then precipitated from the filtrate with oxalate. The hydroxide fraction was purified by dissolving and reprecipitating a number of times and finally was separated by ion-exchange techniques. The alkaline earth fraction was redissolved, purified in the conventional way by scavenging with ferric and lanthanum hydroxides, and the strontium-barium fraction separated in the usual way in strong nitric acid. Strontium and barium were then separated by the chromate procedure. By such techniques strontium-89, yttrium-91, zirconium-95, niobium-95, ruthenium-103, ruthenium-106, rhodium-106, cerium-141, and cerium-144 were identified.

All kinds of marine biota have been collected for radio-assay, including fish, marine invertebrates, algae, and plankton. Usually these have been processed by dry ashing, although some plankton samples were only dried. In general, methods which have been applied to land biota, such as drying and wet or dry ashing, should be adaptable to marine organisms. In the analysis of marine biota there has been greater interest in identifying individual radionuclides, due to the specificity of organisms to extract certain elements from the water.

As demonstrated by biological surveys in the Pacific, radiostrontium does not accumulate in the edible portion of fish or shellfish, with the exception of certain crabs that subsist largely on land vegetation. Radiocesium was not particularly prominent in these organisms. However, a large amount of non-fission-product activity, including isotopes of zinc, cobalt, manganese, and iron, has been found. Conventional chemical methods have been used to separate some elements, and gamma spectroscopy also has been a most valuable tool for radionuclide analyses. Many isotopes, including molybdenum-99, ruthenium-103, ruthenium-106, iodine-131, barium-lanthanum-140, zirconium-95, cerium-141, cerium-144, zinc-65, and cobalt-60, have been identified in plankton.

Relatively little is known of the uptake of radioactive isotopes by marine biota. The most active groups in this field have been the Fishery Radiobiological Laboratory, U.S. Fish and

Wildlife Service, Beaufort, North Carolina, and the Hawaii Marine Laboratory, University of Hawaii, Honolulu.

The North Carolina group has studied the uptake of various isotopes by algae. When 12 species of algae were exposed to a mixture of strontium and yttrium, one species concentrated strontium alone, another equal quantities of both isotopes, and the remainder mostly yttrium, adsorption being the mode of uptake for yttrium. Strontium in the cells of algae was not removed by suspension in non-radioactive water in the absence of a chelating agent such as versene. Of nine species of algae, none took up much cesium-137. Both zinc-65 and ruthenium-106 were taken up from water to a very large extent by *Nitzschia*, which was the only test organism used with these radionuclides.

The Fishery Radiobiological Laboratory also studied the removals of strontium and yttrium from sea water by shrimp, crabs, oysters, clams, scallops, and fish. Except in shells of mollusks, the concentrations in the organisms were not significantly higher than those in the water. In other experiments, mostly with mollusks, cesium-137 was concentrated slightly, and zinc-65 was concentrated very substantially. Ruthenium activity appeared in the digestive tract, with very little being absorbed. Strontium was concentrated by very young flounders to a small extent, appearing chiefly in the gills, bone, and skin. Approximately 27 per cent of the zinc-65 fed remained in fish after 12 hours and was distributed rather widely through the tissues, the largest amounts being in muscle and bone and the highest concentrations in liver and kidney. Cesium was concentrated by a factor of roughly 10 over its concentration in the sea water. Ruthenium, while taken up in the digestive tract, did not pass into the tissues of the fish.

The Hawaii Marine Laboratory has studied the uptake of strontium in fish. Strontium given in individual doses orally is excreted rather rapidly. When fish are maintained in contact with strontium over a long period of time, the concentration in the tissue levels off at a value 0.3 times that in the water, showing that fish are able to exclude strontium ion. The strontium retained is strongly concentrated in the skeleton, gills, and integument.

Another means of establishing uptake of various radionuclides is to compare the

concentration of the stable element in organisms with that in the water. Good data on this subject are extremely meager. Bibliographies on the concentration of selected elements in sea water and in marine biota, whether related to radioactivity or not, are included in Appendix C.

3. ANALYTICAL METHODS

A. Gross Activity of Sea Water

Standard substitute ocean water (ASTM-D-1141-52) has been used in all studies relating to the development of methods for the determination of radioactivity in sea water. The chemical composition of this water is given in Table 4.

Table 4. COMPOSITION OF SIMULATED SEA WATER

Compound	Grams per liter
NaCl	24.53
MgCl ₂	5.20
Na ₂ SO ₄	4.09
CaCl ₂	1.16
KCl	0.695
NaHCO ₃	0.201
KBr	0.101
H ₃ BO ₃	0.027
SrCl ₂	0.025
NaF	0.003
Ba(NO ₃) ₂	0.0000994
Mn(NO ₃) ₂	0.0000340
Cu(NO ₃) ₂	0.0000308
Zn(NO ₃) ₂	0.0000151
Pb(NO ₃) ₂	0.0000066
AgNO ₃	0.00000049

To obtain the gross beta activity of a large volume of sea water, two preliminary tasks must be done: (1) concentrate the active material; and (2) discard the major inert cations present, namely, sodium, magnesium, potassium, and calcium. These could be accomplished by precipitating carbonates and hydroxides by the addition of sodium carbonate, thus eliminating the sodium and potassium, separating the strontium and barium fraction from the carbonate precipitate by the insolubility of the nitrates in strong nitric acid solutions, and separating the radioactive materials from the calcium magnesium nitric acid solution by solvent extraction. Unfortunately, cerium could not be extracted satisfactorily from any such solutions, although yttrium could be. This method of attack was abandoned.

On the basis of the chemical nature of the elements considered important, precipitation of sulfides from basic solution was tried. This should give a precipitate containing all elements with insoluble sulfides, either in acid or basic media, and any elements with insoluble hydroxides. Of the metallic elements, only the alkaline earth metals and the arsenic group would be left in solution by such a procedure. A number of non-metals such as phosphate phosphorus, selenium, and tellurium would also be collected in this precipitate. Strontium and barium could be recovered from the supernatant by precipitation as carbonates, followed by the usual nitric acid separation from calcium and magnesium. In this way, "gross activity" made up of two fractions could be obtained--the sulfide fraction and the alkaline earth fraction. A few of the more important nuclides, particularly cesium-137 and iodine-131, would not be included in this determination of gross activity.

The following procedure was developed. To one liter of sea water (the ASTM substitute ocean water was used) are added lanthanum, iron (III), cobalt, zinc, nickel, and strontium carriers. The solution is acidified, boiled to remove carbonate, made basic with sodium hydroxide, and treated with thioacetamide (70 ml of three per cent solution). By hot digestion sulfides are precipitated. Excess thioacetamide is destroyed by heating until no more hydrogen sulfide odor is noticed. The sulfide precipitate can then be collected, washed, and counted. The supernatant from the sulfide precipitation is made strongly basic with sodium hydroxide and treated with sodium carbonate to precipitate the alkaline earths.

These precipitates are collected, washed, and dried before separation of the strontium-barium fraction from the calcium and magnesium. The efficiency of this method was tested by adding tracer, dissolving the precipitate in acid, diluting to 100 ml, and determining the characteristic peak counting rates on a gamma spectrometer. More than 99 per cent of zirconium-95, niobium-95, ruthenium-106, and cerium-144, are recovered in the basic sulfide fraction, and about 90 per cent of strontium-85 is found in the carbonate precipitate. The strontium was not separated from the calcium-magnesium fraction in this experiment; it is estimated that further losses of about five per cent would be suffered in this separation. The method is to be further tested with chromium-51, cobalt-60, copper-64, zinc-65, and neodymium-147.

B. Radionuclide Analyses in Sea Water

Most of the effort has been concentrated on strontium and is applicable to the determination of either strontium-90 or total radiostrontium.

The first problem considered was the concentration of the strontium from the water sample. Carbonate precipitation has commonly been used for this, usually with pH adjustment to keep the less insoluble magnesium carbonate in solution. It was found that adjustment with an ammonium ion ammonium hydroxide buffer to about pH 8-9 resulted in too much strontium being left unprecipitated. Because of this, a carbonate precipitation in the presence of sodium, rather than of ammonium, hydroxide was chosen. This precipitates strontium, calcium, and presumably most of the magnesium, leaving only two to five per cent of the strontium in solution. In a variation of this method most of the magnesium is first removed from the solution by precipitation with 8-hydroxyquinoline at pH 7. The filtrate is made basic with excess sodium hydroxide and sodium carbonate is added to precipitate the strontium and calcium. The loss of strontium in the magnesium precipitate is only about two per cent. By this technique a much smaller carbonate precipitate is obtained, permitting a better separation of strontium from calcium.

The next phase of the problem to be considered was separation of strontium from calcium. The usual method is precipitation of strontium nitrate from 80 per cent nitric acid. However, on the basis of the work of Sunderman

and Meinke, and also previous experience in this laboratory, it was felt that better removal of calcium could be obtained in 65 to 70 per cent nitric acid. Besides giving definitely superior results, this eliminates use of fuming nitric acid which is a rather nasty reagent. Accordingly, a separation based on treatment of the carbonate precipitate with 30 ml of concentrated nitric acid was tried. The precipitated strontium nitrate was collected and again treated with 30 ml of concentrated nitric acid for a second calcium separation. The first recoveries of strontium by this method were extremely poor. It was suspected that this inefficiency was due to dilution of the nitric acid by the rather large quantities of water retained in the bulky carbonate precipitate. Modification of the method to dry this precipitate in the oven or by an acetone wash proved satisfactory, the loss of strontium in the nitric acid washes being approximately five per cent. The strontium nitrate is taken up in distilled water and purified by separating the barium as chromate and scavenging with iron hydroxide. Total radiostrontium is measured by direct counting, or strontium-90 may be measured by separating yttrium-90 after ingrowth.

In triplicate experiments, using the 3-step procedure, (1) precipitation of magnesium with hydroxyquinoline; (2) precipitation of carbonates in the presence of excess sodium hydroxide; and (3) separation of calcium and strontium by concentrated nitric acid from the dried carbonate precipitate, recoveries of 88.5, 91.3, and 90.0 per cent were obtained, based on gamma counting of tracer strontium-85. The carbonate precipitates contained an average of 65 mg of impurity, which is not felt to be a serious contamination. Where even this much contaminant is a problem, a further purification of strontium using potassium rhodizonate as suggested by Weiss and Shipman can be used. The nitrate precipitate remaining after the first (or second) treatment with nitric acid can be taken up in water, neutralized with ammonium hydroxide, and strontium rhodizonate precipitated. This must then be ignited to carbonate, taken up in dilute acid, and heated. On addition of ammonium hydroxide, a small gelatinous precipitate and some residual carbon separate and are discarded. Strontium carbonate is precipitated from the filtrate by addition of sodium carbonate. This gives a strontium carbonate precipitate of approximately 96 per cent purity with an additional chemical loss in the neighborhood of five per cent. However, it

is felt that this procedure involves too many steps, so it is used only if exceptional purity of the strontium carbonate is required.

Most nuclides of interest other than the strontium isotopes can be determined most simply by gamma spectroscopy. The details of the determinations will vary with the level and with the particular isotope. In the simplest case, where levels are sufficiently high, moderately large volumes of one to four liters of sea water may be examined directly with a gamma scintillation spectrometer. By acidification and boiling, sea water samples may be concentrated approximately ten-fold without separation of salts. The resulting brine may be examined on the gamma scintillation spectrometer with somewhat improved efficiency. Where still further concentration is required, precipitation of carbonate from alkaline solution or of basic sulfides as described for gross counting may give a precipitate suitable for examination by gamma spectroscopy.

C. Radioassay of Marine Biota

These studies have been concentrated on shellfish, specifically oysters and clams. With the continuous ashing device developed at the Center in connection with a milk assay program, as much as one pint (approximately 520g) of the edible portion of such shellfish has been conveniently converted to ash varying from about 3.5 to 7 g. For specific radionuclide determination, this ash can be examined directly by gamma spectroscopy or processed chemically. The separation of the ash into a number of fractions appears to be complicated by the presence of certain anions, probably phosphate.

D. Biological Uptake

It has not appeared feasible for the present to conduct biological uptake studies at the Center because of the difficulty of providing at this inland location a reasonable approximation to the natural environment. As an alternate method of determining uptake is to study the concentration of stable elements in marine biota in comparison to the level of the same elements in the water in which they live. Two methods have been considered for making the measurements in such a study--activation analysis and emission spectroscopy. An evaluation of these two methods indicates that spectroscopy will probably be more economical for routine work,

while activation analysis will permit determination of some of the less common trace elements and also be useful as a check on the spectroscopic results.

One limitation imposed on activation analysis is the high levels of radioactivity which would be produced by the irradiation of sodium in sea water. Preliminary calculations indicate that overnight irradiation of one g of sodium would produce 100 mc of sodium-24 activity. Such samples would have to be processed in a high level laboratory, as at Oak Ridge. The sodium content of sea water can be reduced by saturating with gaseous hydrogen chloride. By evaporating a liter of sea water to approximately 100 ml, and saturating with hydrogen chloride gas in a salt-and-ice bath, sodium chloride was precipitated. Approximately 85 per cent of the sodium chloride can be eliminated in a pure state by this technique.

4. RESULTS

No analyses of sea water or estuarine water have been made to date. A few samples of commercially available oysters and clams obtained from Cincinnati markets have been ashed and subjected to gamma analysis. The ash from these shellfish has shown, in addition to the expected potassium activity, some gamma peaks of extremely low intensity. In the oysters a zinc-65 peak has been identified, along with good indications of ruthenium-106 or ruthenium-103. In addition there were very small peaks at about 0.76 and 0.90 Mev, which have not been identified. The clam sample showed a very prominent peak at 0.5 Mev attributed to ruthenium-103 and a smaller but clear peak at 0.76 Mev which is attributed to zirconium-niobium-95. In addition there was a peak at 1.1 Mev (probably zinc-65) and a rather large peak at low energy which is probably due to cerium-141. Other peaks at 0.94 and 0.22 Mev (probable) have not been identified. This clam sample was obtained in early May and may have shown the effects of recent Soviet nuclear weapons tests.

A preliminary summary report has been prepared by the New York Regional Office of the Public Health Service on coastal water uses along the New England and North Atlantic States. In selecting those areas which may be utilized for the disposal of radioactive waste, the intent is to locate areas in-shore to avoid the need for carrying the radioactive waste

materials in packaged form out beyond the continental shelf, where depths range to 1,000 fathoms, as recommended by the National Committee on Radiation Protection in NBS Handbook 58.

Similar information is being obtained along our Southeastern, Southern, and Western coast lines.

IV. FATE OF RADIOACTIVE MATERIAL

George W. Frishkorn and Richard J. Velten, Chemists, and Helen H. Russell,
Physical Science Aide.

The purpose of this project is to determine (1) factors operative in the partition of specific soluble or insoluble radionuclides into the environment from liquid radioactive waste discharged into streams; and (2) the parameters affecting the utilization of streams for the disposal of radioactive waste.

In the initial phases, areas selected for study will be those where radioactive liquid wastes have been discharged over a period of time during which equilibrium conditions have been approached. The concentration of specific radionuclides, including strontium, cesium, iodine, tritium, and possibly plutonium, will be determined at various distances from the point of discharge.

The results obtained will direct attention to the need to examine other environmental media (biological organisms, vegetation, and bottom muds) for these nuclides. Concentration factors in biological life will be determined, as will the permanency of storage of such radioactivity by the various environmental media under consideration.

By way of initiating this project, a preliminary meeting was held with the Health Physics group at the Knolls Atomic Power Laboratory to arrange for the receipt of samples of their mixed effluent. During March a sample was submitted by KAPL to our laboratory for analysis.

The sample was separated into suspended and dissolved fractions and analyzed for gross alpha, gross beta, strontium-90, total strontium, and barium-140 activity, and gamma spectra of the suspended and dissolved fractions were obtained. The results of the analyses are given in Table 5, and the gamma scans are shown in Figures 13 and 14.

The separation of suspended and dissolved fractions was made by filtration through Whatman No. 41 paper which was then ashed. For radionuclide analysis the suspended solids,

after ashing, were leached four times with boiling 6 N HCl. There was a small amount of residue from this treatment which contained about 1 per cent of the activity. This hydrochloric acid leach was then processed by the usual methods used on dissolved solids. Sr^{90} was determined by purification of the strontium fraction to permit the ingrowth of yttrium for 2 weeks and extraction of this yttrium into TTA followed by back-extracting with nitric acid. Sr^{89} was determined by precipitating and counting the total strontium after extraction of the Y^{90} and subtracting the Sr^{90} value from the total strontium value. Ba^{140} was determined by counting the barium chromate precipitate obtained during purification of the strontium fractions. The barium chromate precipitate from the dissolved fraction was followed for decay. About 1/3 of the activity decayed with the approximate 12-day half-life characteristic of Ba^{140} while 2/3 had a slower decay rate. The barium-140 figure in the dissolved fraction is calculated on the basis of only the 1/3 which has the correct decay rate. Since no decay measurements were made of the Ba^{140} fraction in the suspended solids, it is not known what percentage of that activity was actually due to Ba^{140} ; therefore, the value of 420 $\mu\text{C}/\text{l}$ given in Table 5 must be considered as an upper limit.

The gamma spectra showed only 2 major activities, corresponding to Cs^{137} and to Ce. It is not possible from the energy alone to determine whether the latter was Ce^{141} or Ce^{144} . The spectra also show a very small peak in the neighborhood of 0.5 Mev, presumably due to ruthenium. No other activities could be identified definitely from the gamma spectra.

With the advent of more favorable weather conditions, further KAPL samples will be received for analysis. In the initial phases the purpose of these analyses will be to determine the levels of specific radionuclides in the KAPL waste and to trace these activities downstream as far as possible.

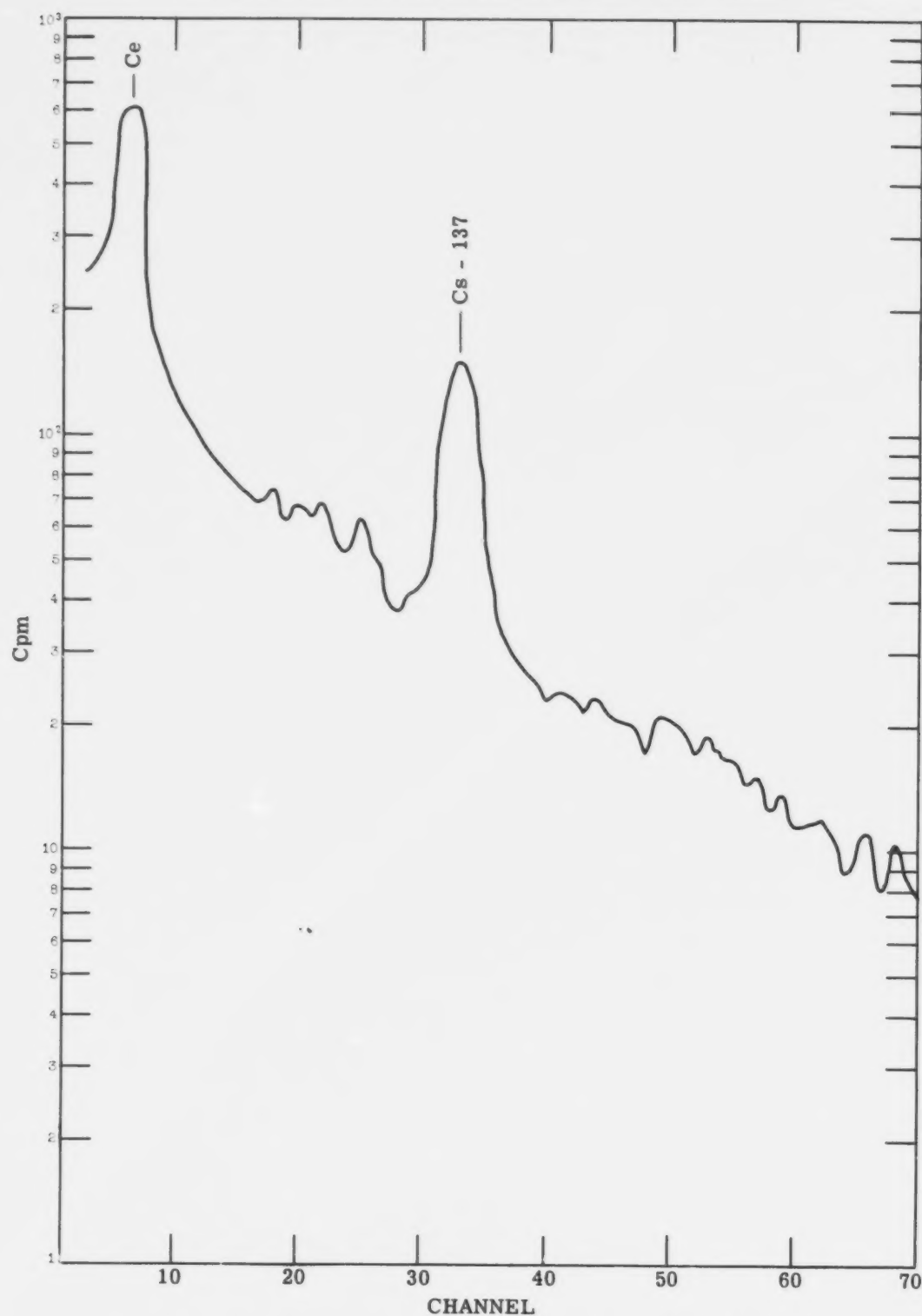


Figure 13. GAMMA SCAN - SUSPENDED SOLIDS - KAPL EFFLUENT
SAMPLE X-80A

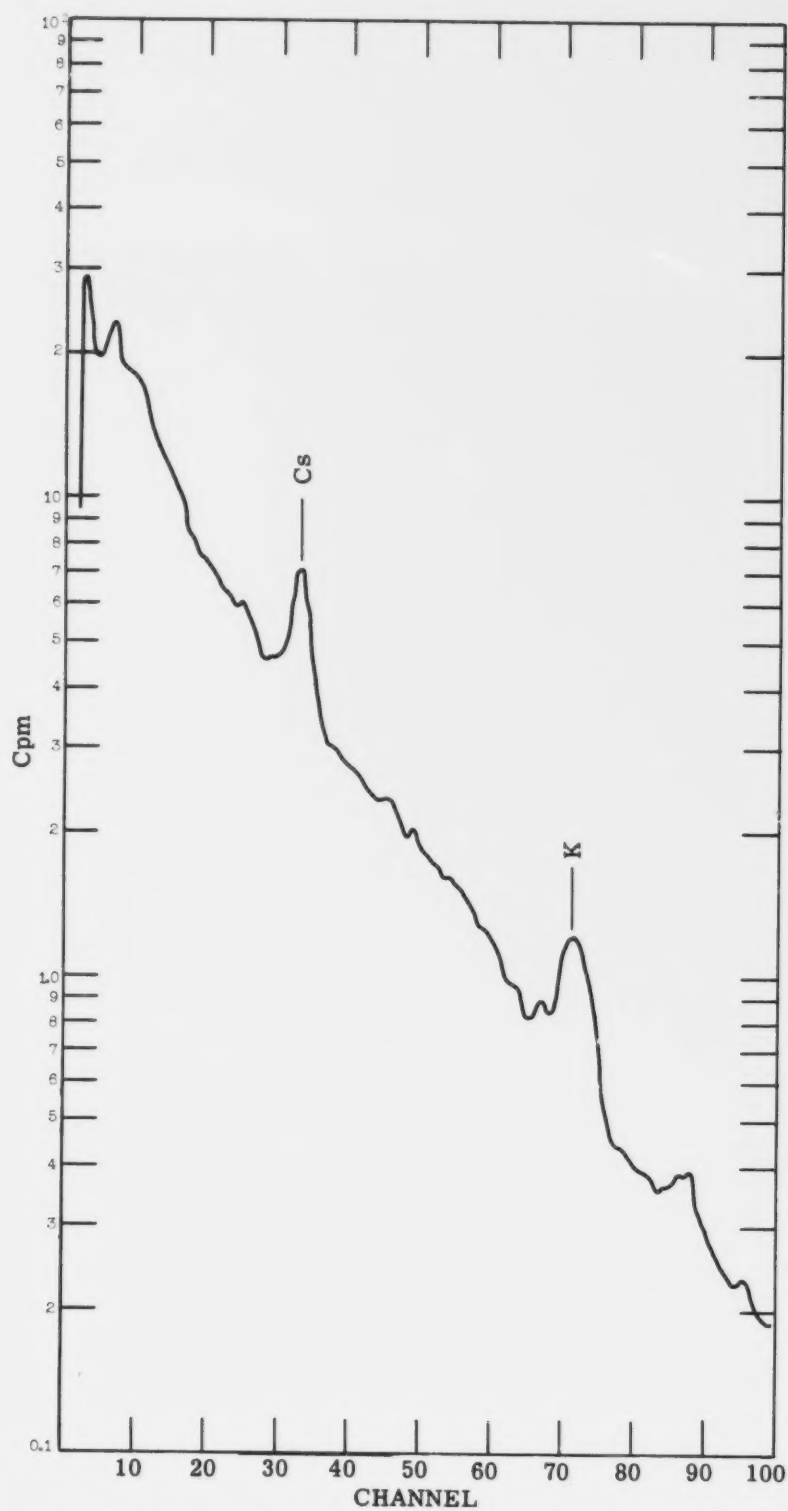


Figure 14. GAMMA SCAN - DISSOLVED SOLIDS - KAPL EFFLUENT
SAMPLE X-80B

Table 5. ACTIVITY OF SAMPLE X-80, KAPL EFFLUENT

 $(\mu\mu\text{c/liter})$

Type	In suspended portion	In dissolved portion	Total
Gross alpha	---	33 ^a	---
Gross beta	15, 500	34, 200	49, 700
Strontium-90	100	12, 400	12, 500
Sirontium-89 ^b	20	1, 600	1, 600
Barium-140	$\leq 420^c$	1, 500	1, 900

a - Assuming 45 per cent counting efficiency

b - Total radiostrontium minus strontium-90

c - This corresponds to total activity of the BaCrO_4 precipitate, including contaminating activities.

APPENDIXES

- A. Tritium Counting - A Review
- B. Location of Important Fisheries in
P. H. S. Regions I and II
- C. Bibliography on the Concentration of
Selected Elements in Sea Water and
in Marine Biota

APPENDIX A

TRITIUM COUNTING—A REVIEW

Seymour Gold, Chemist

1. INTRODUCTION

When charged atomic or nuclear particles pass through matter, the molecules of the material are ionized and excited. The measurement of these ionized particles is the basis for most radiation detection instruments. Uncharged radiation such as X-rays, gamma-rays and neutrons produce secondary ionization which can be measured.

Detection instruments differ in the type of material in which the ionizations occur and in the way in which they are observed. If the particle passes through a gas in a chamber with an electric field across the chamber, the ions move toward the walls and a current is produced. If the charge is relatively small, the primary ions are collected and the chamber is known as an ionization chamber.

The charge can be increased so that the primary ions formed are accelerated and produce secondary ions which also produce further ion multiplication before collection. Under proper conditions, the secondary pulses are proportional to the primary ionizations produced and are therefore proportional to the energy of the radiation depending on the type of radiation. This type of counting is known as proportional counting, or it may be said that the chamber is operating in the proportional region.

If the charge is further increased, a single ion pair in the ion chamber will cause an "avalanche" of ions to form and so cause a large current pulse which is independent of the primary ionizations. The type of instrument which uses this principle is known as a Geiger-Müller or Geiger counter. It may be said that the chamber is operating in the Geiger region.

If the medium through which the ionizing particle passes is super-saturated with a vapor, under proper conditions the vapor will condense

in small droplets along the path of travel of the particle. This phenomenon was utilized as the basis for the cloud chamber which is used for visual observation of nuclear tracks. Fireman and Schwarzer (1) used this type of instrument to count tritium. The disintegrations were counted visually.

It has been discovered that certain materials known as phosphors have the ability to convert the energy of ionizing radiation into visible light photons. If the nuclear or atomic particle passes through a liquid or solid medium containing these phosphoric materials, a pulse of light will be emitted when the particle collides with the phosphor molecule. Counting these flashes or scintillations is the basis of scintillation counters. The phosphor can be in a solid material such as a transparent sodium iodide or a transparent plastic crystal, or in a liquid. The latter is the principle used in liquid scintillation counting.

2. LIQUID SCINTILLATION COUNTING

The fact that radiations will produce scintillations in various materials has been known for some time. Use has been made of this fact by putting radium in various metallic sulfides for use on phosphorescent or luminous watch dials. Zinc sulfide screens have been used to detect alpha particles by counting pulses of light emitted when an alpha particle hits a screen. It was soon learned that common naphthalene would produce scintillations when bombarded by radiation. In 1948 Taschek and Gettings (2) observed that scintillations were produced by tritium beta rays in naphthalene.

A liquid scintillating fission solvent consisting of a solvent and a phosphor is mixed directly with the sample. Since this system offers 4 π geometry for large amounts of sample, the sensitivity for weak beta emitting isotopes is excellent. For this reason

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considerable effort has been exerted in developing methods for counting tritium and carbon-14 by liquid scintillation counting. In 1952, low energy beta rays from tritium and carbon-14 were counted by dissolving labeled compounds in a 0.3 per cent solution of PPO (2,5-diphenyloxazole) in toluene (3). Two photomultiplier tubes at right angles to each other were used to view the pulses. For counting water samples, dioxane was used as the solvent (4). The sample consisted of the dioxane solvent, 0.5 per cent *p*-terphenyl, and 5 per cent water by weight.

A liquid scintillation spectrometer with a fast coincidence circuit which gave 0.06 microseconds resolving time was described in 1953 for measuring tritium and carbon-14 (5). Using this fast coincidence counter, Hayes and Gould (6), counted tritium labeled water and reported a counting efficiency of approximately 6 per cent; but later it was reported (5) that the counting efficiency was actually 10 to 15 per cent. An ion chamber and a vibrating reed electrometer were used to check the results. The scintillation medium consisted of 20 ml of xylene, 100 ml of PPO, and 10 ml of 90 per cent alcohol containing the tritiated water sample. Another use of dioxane was reported using 3 per cent by weight of tritiated water in an 0.5 per cent solution of terphenyl in dioxane (7). The efficiency was reported as 3.8 per cent. In normal operation of the photomultiplier tubes, electrons coming off the cathode have approximately the same energy as pulses produced by tritium and other weak beta particles. These electrons produce what is known as a dark current background which is extremely high in normal operating conditions. The use of a freezer to cool the photomultiplier tubes was reported (8) and said to lower the background in liquid scintillation counting. It was also observed that the light output could be increased 20 to 30 per cent by thorough flushing of the liquid scintillation solution with nitrogen. Complete internal-sample liquid scintillation counting instruments were first made commercially available in 1954. Since that time, reports on liquid scintillation counting have been more numerous. Liquid scintillation counting methods are generally similar, with variations to improve or increase the light output or counting efficiency. A commercially available liquid scintillation unit consists of a freezer chest or some other type of cooling unit

to lower the temperature of the photomultiplier tubes. Two photomultiplier tubes are used in a coincidence counting circuit to reduce greatly the counting of the dark current "noise" inherent in the tubes. A light shield containing a shutter surrounds the photomultiplier tubes and protects them from light while samples are changed. Most variations in liquid counting techniques are variations involving type of solvent, the type of phosphor, and the type of sample. The basic liquid scintillation medium consists of a solvent, a phosphor, and the sample. The best solvents are aromatic liquids such as toluene, xylene, and triethyl-benzene, but these aromatic solvents cannot be used alone for counting water samples, as they are not miscible with water. Therefore, additional solvents are used to dissolve the water in these solvents. One part of water is used with 9 parts of absolute alcohol to produce 90 per cent ethanol, which is soluble in toluene. Another alternative is to use a water soluble solvent as the base for the scintillating solution. Dioxane is often used for this purpose, although it is not as good a scintillating solvent as toluene.

The best phosphors are usually terphenyl or PPO (diphenyloxazole). The purpose of the phosphor is to absorb the energy of the radiation and emit it as a pulse of light. Frequently the pulse of light is not emitted at the wave length at which the photomultiplier tube is most sensitive. For this reason wave length shifters are added to the scintillation medium to absorb the light emitted by the phosphor and re-emit it at a wave length closest to the photo tube sensitivity peak. The most commonly used wave length shifter is POPOP (1, 4 - di [2 - (5-phenyloxazolyl)] benzene).

Other solvents, phosphors, and wave length shifters in various combinations and concentrations have been used and described. Counting efficiencies are usually low, in the order of 1 to 10 per cent. For high activity counting, as where tracer materials are used, almost any scintillation solvent system may be employed. With low level activities a system holding a large volume of sample and having a high efficiency is desirable. The figure of merit of the scintillating solution, that is, the product of the counting efficiency and the volume fraction of sample contained in the solution, multiplied by 10,000, gives a relative efficacy of the system under study.*

* See page 10 for example where figure of merit is calculated.

One system described (9) consists of 2.3 per cent water, 34.5 per cent ethanol, and 63.2 per cent toluene. The counting efficiency is about 4.3 per cent. This gives a figure of merit of 2.3×4.3 or 10. The phosphors used are PPO and POPOP. It was found that the use of naphthalene increases the counting efficiency, since naphthalene is solution with the solvent forms a new scintillation solvent with a much better energy transfer property (10)(11). To assay tritium in body fluids, Langham et al (12) used a solvent consisting of 1 liter dioxane, 7 grams PPO, 50 mg POPOP, and 50 grams of naphthalene. To 24 ml of this solvent, 1 ml of water was added. The counting efficiency was reported to be from 6 to 9 per cent. Since the sample contained 4 per cent water, we can calculate a figure of merit of approximately 30. Another solvent system (13) consisted of 2 ml of water sample, 20 ml of ethanol, 28 ml of toluene, and 100 mg of PPO. This gave a 4 per cent solution of water with a counting efficiency of about 3 per cent for a figure of merit of about 12. When preparing the samples for counting, 0.1 ml of distilled water was added to each sample for a total of 2.1 ml water. In a similar sample 0.1 ml of tritiated water sample was added. Using this internal standard, the counting efficiency for each sample could be determined separately. This was necessary since the quenching effect of urine varies from sample to sample. This method is very useful when counting samples of variable purity and color. Quenching occurs when certain impurities in the scintillation medium absorb the energy from the phosphors and convert it to heat. A complex system made up of 5 parts xylene, 5 parts dioxane; 3 parts ethanol; and 1 part water gave a counting efficiency of 4.2 per cent. Since the water amounted to 7.7 per cent, this gave a figure of merit of 32. The solutes consisted of 80 grams liter of naphthalene, 5 g/l PPO, and 0.05 g/l alpha NPO (α -naphthylphenyloxazole) (14). One of the best solvent systems yet reported for water samples (15), consists of dioxane, anisole, and dimethoxyethane in the ratio of 6 : 1 : 1 by volume. To 800 ml of this mixture, 12 grams of PPO and 0.5 gram POPOP is added. Enough of this solvent is taken and mixed with the water sample to produce a 20 per cent water solution. The counting efficiency is approximately 4 per cent, giving a figure of merit of 80. Since reagent grade chemicals result in a low counting efficiency it is important that the solvents be rigorously purified. This solvent system is referred to as 611 or Polyether - 611 solvent.

Laundry detergent "whiteners" have been examined as a new source of scintillation phosphors (16). Further investigation will be necessary. Isopropanol has been reported to give higher counting efficiencies than ethanol or methanol when used with water and toluene mixtures (17).

For a time there was much confusion as to which phosphors and wave length shifters form the best combination. Hayes and co-workers (18) reported that POPOP and terphenyl in the ratio of 1 : 40 were better than POPOP and PPO. However, Avivi (19) later reported that PPO and POPOP in toluene was the best mixture. Some of this confusion was recently cleared up when it was shown that all photomultiplier tubes did not have the same sensitivity (20). The relative frequency output of the scintillating solution should match the photomultiplier frequency response. For a photomultiplier tube with a maximum sensitivity at 4440 angstroms, POPOP would be the best wave length shifter. For a photomultiplier tube with a maximum sensitivity at 3600 angstroms, the best wave length shifter would be terphenyl. In this case terphenyl would be used alone. Therefore, different workers using the same scintillating solvent mixtures would obtain different results if their photomultiplier tubes differed in spectral sensitivity. Although the manufacturer claims a certain photocade sensitivity for the photomultiplier tubes, there are normal variations in the manufacturing process which will cause the peak sensitivity to differ from published values. It is recommended that the spectral sensitivity of the photomultiplier tubes be determined individually in order that the most efficient wave length shifter can be used with each installation.

In an attempt to avoid the use of water in liquid scintillation media, various schemes have been investigated to convert the water to an organic material which will be soluble in toluene. Leger (21) converts water to paraldehyde which is soluble in the scintillation liquid. Acetylene was converted to paraldehyde by the use of Nieuwland's catalyst (22).

Another scheme was the conversion of tritiated water to tritiated toluene by an exchange procedure using sulphuric acid as a catalyst (23). In this method, tritiated water is shaken for about 120 hr with fuming sulphuric acid and toluene. The mixture is cooled, and the toluene is decanted. About 30 per cent of the

tritium from the water is exchanged with the toluene. This method appears very useful since the toluene produced can be used directly as the scintillation solvent.

Other methods have been reported for counting solid samples in the scintillation counter (24). In this procedure any solid sample insoluble in toluene can be used. The sample must be ground into very fine powder to form a stable suspension and to minimize self-absorption. The various materials used include barium-carbonate containing carbon-14, sodium acetate containing tritium, barium sulphate containing sulfur-35, calcium oxalate and calcium carbonate containing calcium-45, various other tissues, and bacteria. The pulverized sample is placed in a vial containing the liquid scintillation medium and shaken. The suspension is counted in the liquid scintillation counter. Among the problems to be evaluated are settling of the suspension, self-absorption, and light-absorption. These problems have been solved to some extent. Two papers appearing about the same time (25)(26) make use of thickening agents to form a gel in which the suspension is held. This gel keeps the particles from settling out and solves one of the major problems associated with suspension counting. In one case (25) a gel called thixcin is blended with the sample, PPO, and the solvent in a blender. It was reported that no self-absorption by the particles was found but that the optical properties are affected by the suspension. Smaller bottles with narrower absorbing pads gave higher counting rates (25). In the other case (26), aluminum stearate was used as a thickening agent. PPO and POPOP in xylene or toluene were used with the aluminum stearate.

Oxygen gas has been reported as a significant light quencher. Therefore, it has been advocated that oxygen be removed from the liquid scintillation sample before counting. The use of an ultrasonic degasser (27) made the removal of oxygen a simple problem.

Another problem arises from the fact that potassium-40 in the glass vials also produces light pulses which are picked up by the photomultiplier tubes. The use of silica vials (28) lowered the background from 70 cpm with a glass vial to 17 cpm. Vicor vials gave a back-

ground of 40 cpm in the tritium range.

Since the commercial liquid scintillation instruments are equipped with two channels, the instrument can be set up to count two isotopes simultaneously. Tritium and carbon-14 levels may be determined in the same sample by adjusting the various discriminator settings and measuring the tritium to carbon-14 ratio in the various channels (29).

For high activity work, simple single photomultiplier tube scintillation counters have been built. The output of the photomultiplier tube is connected to a pre-amplifier and the output pulse is then received by a scaler (30). However, it was found that initial noise was so high that carbon-14 or tritium counting was impossible (31). After a few months the noise level in the tritium range dropped considerably. It was found that the glass in the photo tube fluoresced strongly after exposure to bright light. This fluorescence decayed very slowly. It was recommended that photomultiplier tubes used for soft beta counting should never be exposed to strong light after purchase from the manufacturer. A single photomultiplier tube scintillation counter is inexpensive and is quite simple. No refrigeration is used, but, because of the higher dark current background, samples of high activity level only can be counted (32).

An even simpler device makes use of a galvanometer sufficiently sensitive so that it can be connected directly to the output of the photomultiplier tube to measure the current (33). This device appears to be similar to an ion chamber set-up which measures current and not individual pulses. Sensitivity of this instrument was reported to be from 0.035 to 1.45 μC of tritium.

Liquid scintillation counting requires very little if any sample preparation. This is in strong contrast to gas counting methods where complete gas-handling systems must be set up requiring, for example, liquid nitrogen or liquid air traps to purify the gas. Although liquid scintillation methods are not quite as sensitive at the present time as gas counting methods, the ease of operation and time saved makes liquid scintillation counting a more desirable method.

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APPENDIX B

LOCATION OF IMPORTANT FISHERIES IN PHS REGIONS I & II

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1. INTRODUCTION

The purpose of this report is to provide a rough indication of the location of the important marine fisheries in PHS Regions I & II, New York, Pennsylvania, Maine, Vermont, New Hampshire, Massachusetts, Connecticut, Rhode Island, New Jersey, and Delaware. This is to serve as background information for persons working in the radiological health program who may be concerned with the relationship of marine fishery resources to the disposal of radioactive materials into the marine environment.

In view of this purpose an attempt has been made to indicate the "time" factor for the location of a particular marine fish in a particular part of the environment. The information which is presented in this report should be considered as a "crude" first approximation since no attempt is made to pinpoint the fisheries except in general terms.

The information contained herein was taken from the reports of the Fish and Wildlife Service (FWS) and the New England-New York Inter-Agency Committee (NENYIAC).

2. SUMMARY

The entire Continental Shelf area is important as an environment for the commercial fisheries of the United States. The use of this area for disposal of radioactive wastes would have to consider the effect upon the environment of particular species inhabiting the area for a part or all of their life cycle.

Some fish types are more mobile than others. Shellfish are the most restricted and would be affected over their entire life cycle.

Finfish, except for a few species being limited to selected topographical boundaries, are much more mobile.

This report identifies the various species in the area and the possible effect of waste discharges on commercial fishing.

3. MARINE FISHERY TYPES AND ENVIRONMENT

The "time" that a particular fish remains in a given area is dependent first of all on its mobility which may be relatively unlimited as in the case of the "finfish" or practically restricted or non-existent as in the case of the "shellfish". Therefore, the primary divisions for this report will be "finfish" and "shellfish".

A. Finfish Environments

The environmental preferences of finfish may be rather restrictive for the entire life cycle or as is the usual case only during certain periods of spawning with a general temperature dependence related in part to the food resource of the particular fish.

(1) Groundfish

The groundfish are the most important marine resource of New England. As the name implies the term groundfish applies to many kinds of fish that live on or near the bottom. Usually they are confined to depths shallower than 100 to 150 fathoms. Although the "Continental Shelf" determines the outer limits of the groundfish environments there are certain local areas, known as the "Banks", of particular importance as a source of groundfish. The sketch on the top of the next page locates these "Banks" of interest to this study.



Figure B-1 - BANKS WITH AREAS SHOWN IN SQUARE MILES.

The environmental factors for the various groundfish are as follows:

Ocean Perch (Rosefish) - Taken from depths of 50 to 125 fathoms. Large fish move about from one area to another so that there is exchange between "Banks".

Haddock - No interchange between "Banks". An important nursery area exists on southeast Georges Bank at 50 to 60 fathoms.

Cod - Similar to Haddock but with more movement between near-by "Bank's".

Flounders

Yellowtail - 10 to 50 fathoms of water. Spawning along shore areas.

Blackback (winter) - Spawn depths 1-3 fathoms. Relatively nonmigratory. Found outside bays in summer to inside waters along coast in winter. Southern New England and Long Island Sound an important area.

Grey Sole - 25 to 120 fathoms.

Sea Dab - 15 to 60 fathoms.

Lemon Sole - Limited to Georges Bank only.

Halibut - Off Nova Scotia at 100 to 200 fathoms. Coastal grounds have been fished out.

Pollock (Boston Bluefish) - Spawn of Massachusetts Bay October thru February. Taken from Gulf of Maine within 75 fathom contour.

Cusk - Taken from depths greater than 60 fathoms. Spawn during summer. Cape Cod marks southern limit of important areas.

Whiting (Silver Hake) - Taken from Sable Island Bank south to South Carolina from shoreline to depths of 300 fathoms. Gulf of Maine a summer fish. Off Long Island a year around fish, taken offshore in winter and inshore in spring.

Hake (White and Red) - From tide line to depths of 300 fathoms from Newfoundland to Cape Hatteras. Offshore in summer and inshore in winter. Enters harbors in winter.

Tilefish - Outer edge of continental shelf in depths of 50 to 200 fathoms. Limited to outer edge of shelf.

Wolffish - General distribution north of Cape Cod and from tidelines to depths of 500 feet or more.

(2) Shorefish

"Much of the shorefish resource is composed of migratory populations of fish that move from offshore southern wintering grounds to northern inshore grounds in summer." The peculiar migratory habits of the shorefish are demonstrated by the following special environmental factors:

Scup - Taken in inshore waters of New Jersey, New York and southern New England in summer and during winter from the offshore waters of the Jersey Capes southward. Spawn in inshore waters and bays of New Jersey, Long Island and southern New England from May to August. Same grounds serve as nursery grounds.

Butterfish - Spawn in June and July in bays, sounds and inshore waters from Chesapeake Bay to southern New England. Seldom found north of Cape Cod but some found on Georges Bank. Generally found in summer only.

Summer Flounder (Fluke) - Winter fishery off New Jersey. Summer from southern shore of Long Island, and on the coasts of New Jersey and Delaware. Believed to spawn offshore. Range as far as Georges Bank but generally shallow inshore waters.

Shad - Spawn in fresh water in spring. Ocean remainder of the year. All along coast, in this area.

Alewives - Spawn in fresh water in spring. Ocean remainder of year. All along coast in this area.

Tautog (Blackfish) - From Bay of Fundy to South Carolina but most abundant between Cape Cod and Delaware Capes. Move to deeper waters in winter. Spawn in June in southern New England waters.

Sea Bass - Similar to Tautog.

Common Eel - Spawns in ocean and spends most of life in inland waters.

Ocean Pout - From both inshore and from "Bank's".

Striped Bass - Spawns in upper Delaware Bay and lower Hudson River in spring and early summer. Winter in Delaware Bay, Hudson River and other coastal rivers and bays of New

Jersey, Long Island and southern New England. Limited to within 1 to 2 miles from shore. Migrate primarily to New England waters in summer.

(3) Pelagic Fish

These are the free-swimming fishes of the open sea which are not dependent on certain restricted habitats or localities. "These fish follow the seasons, or more correctly, follow their food which is abundant or scarce depending on the season. With the exception of the swordfish and marlins most commercial pelagic fish are found in huge schools which appear and disappear with dramatic suddenness."

Menhaden - In spring large schools appear in coastal waters even brackish waters. Migrate southward in winter to unknown winter spawning grounds. Taken during fall migration.

Herring - Taken from New Jersey thru Maine. Spawn in summer and fall along shore. Schools of small herring appear inshore from spring to fall. Spawn at depths of 12 to 180 feet and as far as 25 miles from shore.

Mackerel - Spawn from Cape Hatteras to Gulf of St. Lawrence with principal areas between Chesapeake Bay and Cape Cod Bay. Appear off New Jersey in April and enter Gulf of Maine in late June and July returning south in September.

Thimble-Eyed Mackerel - Similar to mackerel except seldom appearing north of Cape Cod.

Smelt - Spawn short distances above tidewater. More prevalent in northern New England waters. Summer in estuary or near-by sea. Spend winter in harbors or brackish waters.

Bluefish - Move north in spring and south in fall. Spawn offshore. Come inshore in May or June when young.

Swordfish - Block Island best area for fishery. Close to coast only when of large size. Appear only in summer.

Tuna - Summer only.

(4) Finfish Catch Statistics

For purposes of comparison, the statistical information for the year 1952 is used in this report. The data represent the places

where particular fish were landed and thus may not be a true indicator of the actual grounds where taken. In the case of some of the inshore fish these data are more representative of the importance of the fishery in that area. Following the tabulation of the relative catch for some of the more important fish is a map showing the

areas where fish were taken and landed in Massachusetts. Since Massachusetts is the receiving area for a majority of the fish taken offshore these data should be representative of the relative importance of the particular areas as well as of the types of fish taken.

The following is a comparison of the value and size of the marine fishery in Regions I & II for 1952:

FISH	NEW ENGLAND STATES		MIDDLE ATLANTIC STATES	
	Thousands of Pounds	Thousands of Dollars	Thousands of Pounds	Thousands of Dollars
Ocean Perch	189,042	8,210	-	-
Haddock	161,341	12,491	156	14
Cod	42,401	2,994	1,282	163
Flounders				
Yellowtail	16,744	1,947	241	25
Blackback	14,851	1,986	926	94
Grey Sole	5,748	564	20	1
Sea Dab	7,606	678	35	2
Lemon Sole	3,170	564	14	2
Halibut	395	103	-	-
Pollock	26,953	995	-	-
Cusk	3,699	194	-	-
Whiting	105,954	2,217	2,469	146
Hake				
White	11,783	554	120	7
Red	3,956	72	455	16
Tilefish	1,649	157	466	43
Wolffish	2,009	135	-	-
Scup	6,807	391	18,942	1,311
Butterfish	4,635	401	6,130	445
Summer Flounder	6,860	1,308	8,470	1,554
Shad	578	70	2,240	417
Alewives	5,930	69	212	8
Tautog	130	6	49	2
Sea Bass	860	70	49	2
Common Eel	101	18	238	41
Ocean Pout	404	6	-	-
Striped Bass	161	39	1,141	280
Menhaden	36,087	443	480,304	4,501
Herring	153,513	1,873	1,632	32
Mackerel	12,525	954	2,241	331
Thimble-Eyed Mackerel	115	2	263	29
Smelt	177	41	-	-
Bluefish	139	27	1,648	296
Swordfish	301	112	49	2
Tuna	1,145	142	783	39

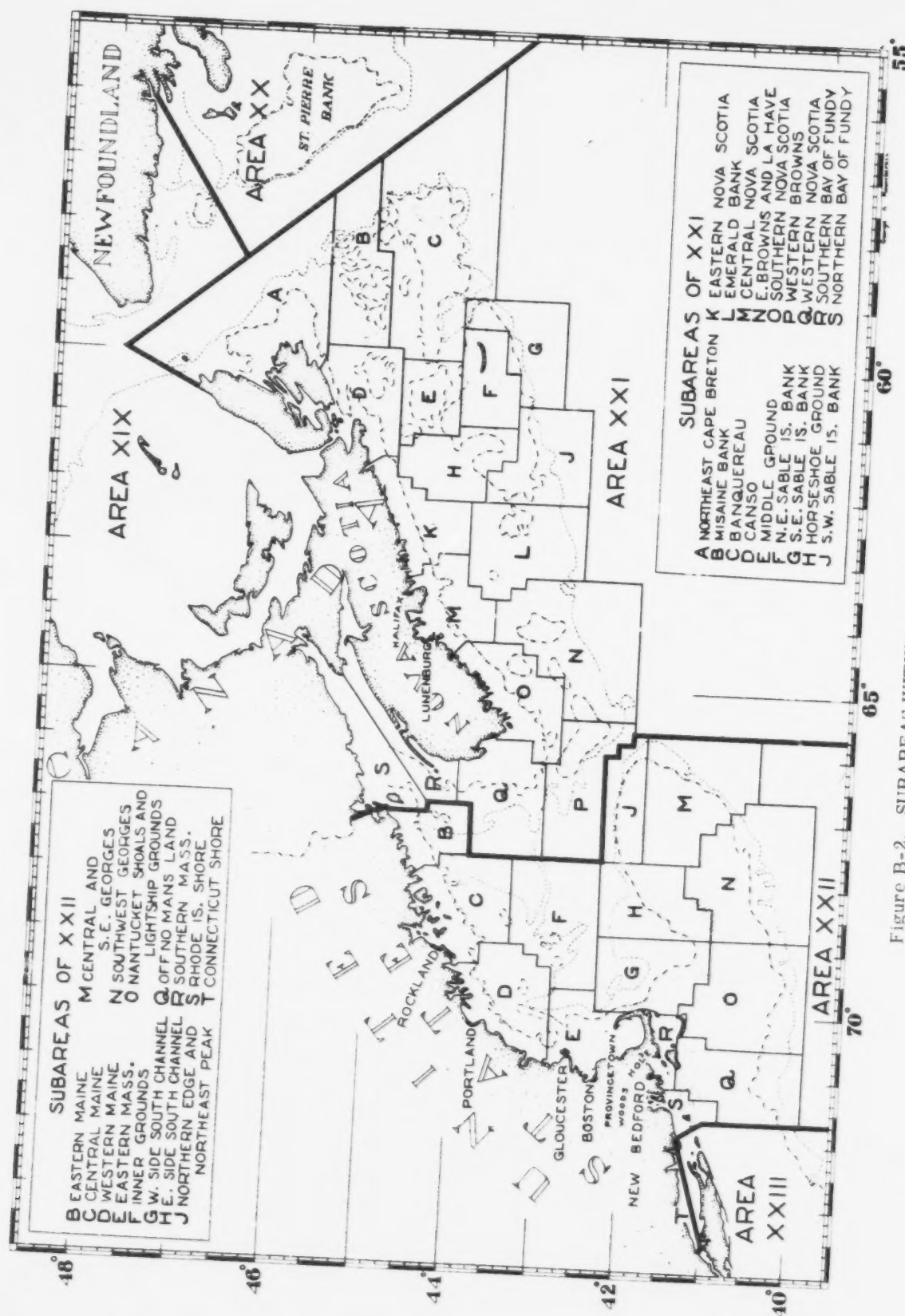


Figure B-2. SUBAREAS WITHIN AREAS XXI & XXII.

Table B-1. SUMMARY OF FISHERY BY AREA AND SUBAREA, 1952.

AREA AND SUBAREA	CHAFT FISHING NUMBER	TRIPS	DAYS ABSENT	COD, DRAWN		LUSK, DRAWN	FLOUNDERS, FOUND	
				LARGE POUNDS	MARKET POUNDS		GRAY SOLE POUNDS	YELLOW- TAIL POUNDS
GULF OF ST. LAWRENCE (AREA XIX):								
OFF ST. LAWRENCE, UNCL	3	57.6	710	24,842	37,940	-	43,472	-
OFF GRAND BANK (AREA XX):								
GRAND BANK	32	182.7	2,289	6,165	33,745	-	25,120	-
ST. PIERRE BANK	1/33	196.6	2,331	8,270	33,745	-	25,775	-
TOTAL							25,895	-
OFF NOVA SCOTIA (AREA XXI):								
NORTHEAST CAPE BRETON	21	29.9	357	50,967	3,900	-	11,318	-
MISMAINE BANK	10	13.4	153	212,917	56,042	-	23,558	-
BANQUEREAU	49	85.9	989	116,877	251,048	1,582	375,922	12,120
CANSO BANK	39	46.1	518	37,034	26,145	4,630	446,277	95
MIDDLE GROUND	42	30.4	302	63,789	104,276	10,008	195,882	13,790
N.E. SABLE ISLAND BANK	36	93.3	839	415,483	1,339,280	683,148	60,591	296,129
S.E. SABLE ISLAND BANK	29	17.6	171	180,625	342,380	174,600	23,760	37,395
HORSERIDGE GROUND	83	179.6	1,995	606,565	581,451	76,080	692,773	14,310
S.W. SABLE ISLAND BANK	42	52.3	554	897,170	690,545	129,565	35,033	19,370
EASTERN NOVA SCOTIA	43	53.5	621	17,471	15,857	13,916	9,380	-
EMERALD BANK	39	25.1	252	80,914	65,426	20,783	9,278	560
CENTRAL NOVA SCOTIA	46	56.3	640	34,825	29,477	5,476	25,399	285
E. BROWN AND LA HAVRE	100	162.8	1,448	693,243	731,568	83,190	85,714	1,782
SOUTHERN NOVA SCOTIA	99	149.7	1,404	168,395	197,632	21,636	25,561	2,500
WESTERN BROWN	59	78.0	661	319,978	430,288	30,156	10,784	2,588
WESTERN NOVA SCOTIA	12	12.3	111	6,580	14,924	1,432	5,413	40,485
SOUTHERN BAY OF FUNDY	2	2.0	16	2,280	3,500	1,700	500	-
NOVA SCOTIA, UNCL	15	16.5	185	27,700	27,625	2,795	61,390	2,100
NORTHERN BAY OF FUNDY	1	.1	1	45	130	-	-	-
TOTAL	1/139	1,104.8	11,217	3,933,148	4,911,394	275,282	2,055,520	441,719
OFF NEW ENGLAND (AREA XXII):								
EASTERN MAINE	15	35.9	182	17,760	85,317	8,380	12,224	-
CENTRAL MAINE	8	12.5	74	5,002	3,768	7,798	23,942	-
WESTERN MAINE	97	937.6	1,923	399,013	376,927	279,540	363,231	20,326
EASTERN MASSACHUSETTS	351	11,278.1	13,292	921,059	1,197,132	550,131	347,210	634,497
INNER GROUNDS	116	546.5	2,734	272,883	233,837	530,924	222,617	1,730
N. GULF OF MAINE, UNCL	13	13.5	98	3,580	4,610	5,130	5,080	100
WESTERN SIDE SOUTH CHANNEL . .	307	1,603.5	8,338	598,596	2,486,380	204,813	362,726	805,758
EASTERN SIDE SOUTH CHANNEL . .	178	542.8	3,798	321,114	599,606	176,727	387,397	676,374
NORTHERN EDGE OF GEORGES . . .	172	1,054.3	8,756	1,117,041	3,821,565	61,395	46,113	193,981
CENTRAL AND S.E. GEORGES	188	694.6	5,493	1,336,022	980,165	11,525	14,162	30,870
SOUTHWEST GEORGES	170	475.6	3,269	1,336,823	433,600	13,575	107,895	1,959,720
NANTUCKET SHOALS & LIGHTSHIP .	169	864.0	3,566	44,323	746,780	250	342,060	4,566,375
OFF NO MAN'S LAND	111	894.4	2,034	4,875	38,530	-	11,943	2,451,930
SOUTHERN MASSACHUSETTS	43	138.0	421	255	13,210	-	25	1,324,371
RHODE ISLAND SHORE	3	3.0	14	-	-	-	-	2,975
S. NEW ENGLAND, UNCL	148	362.8	1,270	86,491	191,285	1,245	38,350	25,365
CAPE COD	409	10,381.0	10,381	961,319	1,126,924	69,189	228,645	342,135
TOTAL	1/1,101	29,828.1	65,043	6,776,156	12,339,806	1,920,622	2,171,560	1,919,969
OFF MIDDLE ATLANTIC STATES (AREA XXIII):								
SOUTH	14	14.9	93	-	-	-	-	125
PACIFIC OCEAN	1	1.0	116	-	-	-	-	-
GRAND TOTAL	1/1,130	31,193.0	80,110	10,742,416	17,322,885	2,195,904	4,296,572	14,746,946

(CONTINUED ON NEXT PAGE)

SEE FOOTNOTES AT END OF TABLE.

Table B-1. SUMMARY OF FISHERY BY AREA AND SUBAREA, 1952. (Cont'd)

AREA AND SUBAREA	FLOUNDERS, ROUND-COULAUED		HADDOCK, DRAWN		HAKE		DRESSED		HALIBUT, DRAWN	MACKEREL, ROUND
	BLACK-BACK POUNDS	DAB POUNDS	FLUKE POUNDS	LARGE POUNDS	SCROD POUNDS	RED, ROUND POUNDS	WHITE, LARGE POUNDS	SMALL POUNDS		
GULF OF ST. LAWRENCE (AREA XIX):										
OFF GRAND BANK (AREA XX):										
ST. PIERRE BANK	-	12,170	-	73,200	38,775	-	35,608	-	420	-
TOTAL	-	150,115	-	3,355	700	-	9,165	-	265	-
OFF NOVA SCOTIA (AREA XXI):										
NORTHEAST CAPE BRETON	-	10,645	-	4,200	1,055	-	6,502	-	96	-
MISKINE BANK	-	1,171	-	8,541	1,401	-	4,624	-	1,740	800
BANQUEREAU	-	35,955	-	101,105	169,750	-	26,658	2,155	321	1,072
CANSO BANK	-	11,296	-	16,874	9,728	-	14,167	3,791	-	-
MIDDLE GROUND	-	20,555	-	970,244	705,981	-	48,885	-	2,404	2,654
N.E. SABLE ISLAND BANK	1,919	209,270	-	2,111,955	4,795,904	-	22,943	-	7,985	36,203
S.E. SABLE ISLAND BANK	960	36,502	-	3,346,875	683,573	-	1,440	1,710	1,346	67
HORSESHOE GROUND	670	45,708	-	6,409,720	2,996,671	-	263,466	34,563	14,508	18,410
S.W. SABLE ISLAND BANK	198	32,540	-	2,466,855	1,943,320	-	26,065	-	14,221	67,698
EMERALD BANK	-	6,025	-	53,597	25,199	-	28,730	5,056	74	48
CENTRAL NOVA SCOTIA	-	4,185	-	459,061	294,772	-	20,057	1,385	1,588	21,165
E. BROWNS AND LA HAVE	-	10,175	-	96,551	24,159	800	36,092	6,050	966	200
SOUTHERN NOVA SCOTIA	580	56,577	-	6,723,896	4,236,421	-	89,975	3,964	13,418	545
WESTERN BROWNS	780	48,021	-	2,157,717	1,221,885	1,740	76,297	16,711	3,391	123
WESTERN NOVA SCOTIA	5,130	37,372	-	3,112,337	2,253,180	-	27,361	800	9,837	1,630
SOUTHERN BAY OF FUNDY	31,475	1,501	-	187,446	212,198	-	2,050	-	73	-
NOVA SCOTIA, UNCL.	5,350	2,915	-	31,000	44,500	-	21,285	1,840	150	-
NORTHERN BAY OF FUNDY	230	-	-	168,535	100,440	-	21,110	-	-	-
TOTAL	47,462	570,503	-	27,426,309	19,724,127	2,540	730,977	78,584	72,118	150,615
OFF NEW ENGLAND (AREA XXII):										
EASTERN MAINE	200	3,026	-	208,167	170,962	-	15,136	1,850	717	-
CENTRAL MAINE	-	17,130	-	9,210	12,843	-	4,077	9,715	20	-
WESTERN MAINE	7,385	192,812	-	1,112,132	1,024,518	205,525	431,961	216,427	2,441	3,530
EASTERN MASSACHUSETTS	381,365	345,973	4,672	1,540,498	931,015	1,353,328	1,352,015	143,250	5,294	5,259,919
INNER GROUNDS	1,350	139,468	-	1,519,696	711,313	1,800	485,218	127,913	4,411	-
OF MAINE, UNCL.	725	4,995	-	33,565	36,695	-	6,045	3,125	-	-
WESTERN SIDE SOUTH CHANNEL	2,394,139	433,091	48,910	4,408,678	6,787,339	84,350	616,413	29,37	12,436	38,030
EASTERN SIDE SOUTH CHANNEL	1,092,228	2,135,656	30,540	4,858,275	5,605,091	2,500	302,223	6,334	17,478	5,395
NORTHERN EDGE OF GEORGES	484,052	229,065	6,300	7,110,714	24,267,715	2,100	217,112	2,350	26,574	167,660
CENTRAL AND S.E. GEORGES	1,031,708	65,092	6,700	3,506,386	9,860,210	-	55,220	-	15,811	12,715
SOUTHWEST GEORGES	426,715	112,025	8,785	3,625,107	4,933,415	-	39,415	1,010	17,272	615
NANTUCKET SHOALS & LIGHTSHIP	2,403,318	25,310	2,127,690	743,907	1,246,402	30	20,440	485	1,275	-
OFF NO MAN'S LAND	207,105	930	905,893	10,400	17,720	51,850	2,530	95	-	10,245
SOUTHERN MASSACHUSETTS	385,175	-	163,353	10,250	-	-	-	-	-	-
RHODE ISLAND SHORE	200	-	1,295	-	-	-	-	-	-	-
S. NEW ENGLAND, UNCL.	517,920	159,200	126,090	406,675	929,395	-	7,810	-	939	9,800
CAPE COD	897,737	871,805	1,286,049	2,521,326	398,571	1,609,812	145,334	35,188	88,687	4,592,788
TOTAL	10,229,322	4,735,578	4,710,324	31,614,986	56,944,939	3,311,295	3,700,949	577,139	193,355	10,100,697
OFF MIDDLE ATLANTIC STATES (AREA XXIII):										
SOUTH	-	-	134,125	-	-	-	125	-	-	-
GRAND TOTAL	10,276,784	5,468,366	4,844,449	59,118,300	76,708,541	3,313,835	4,478,744	655,723	266,156	10,251,312

SEE FOOTNOTES AT END OF TABLE.

Table B-1. SUMMARY OF FISHERY BY AREA AND SUBAREA, 1952. (Cont'd)

AREA AND SUBAREA	OCEAN PERCH, POUNDS	POLLOCK, DRAWN POUNDS	SCUP OR FORGY, ROUND POUNDS	SMOKED-FISH, DRESSED POUNDS	WHITING		WOLFFISH, DRAWN POUNDS	SCALLOPS (MEATS) POUNDS	OTHER, AS LANDED POUNDS	TOTAL, AS LANDED POUNDS
					DRESSED POUNDS	ROUND POUNDS				
GULF OF ST. LAWRENCE (AREA XIX):										
GULF OF ST. LAWRENCE, UNCL.	7,893,075	1,922	-	-	-	-	-	-	-	-
OFF GRAND BANK (AREA XX):										
ST. PIERRE BANK.	33,747,006	4,700	-	-	-	-	385	-	87,185	34,069,906
TOTAL.	556,000	3,280	-	-	-	-	385	-	8,000	572,530
OFF NOVA SCOTIA (AREA XXI):										
NORTHEAST CAPE BRETON.	34,305,006	7,990	-	-	-	-	-	-	95,185	34,642,436
MISATINE BANK.	4,234,310	1,708	-	-	-	-	-	-	65,400	4,390,001
BANQUEREAU.	1,310,675	14,835	-	-	-	-	229	-	18,600	1,654,975
CANSO BANK.	10,598,123	119,696	-	-	-	-	4,242	-	291,460	12,379,515
MIDDLE GROUND.	4,813,248	57,366	-	-	173,500	-	1,902	-	133,570	5,576,444
N.E. SABLE ISLAND BANK.	804,230	218,769	-	-	-	-	22,227	-	7,820	3,234,671
S.E. SABLE ISLAND BANK.	69,220	485,990	-	-	-	-	63,270	-	560	10,621,654
MORSE SPRUCE GROUND.	283,150	111,638	-	13,998	-	-	11,430	-	57,375	2,295,798
S.W. SABLE ISLAND BANK.	8,110,978	1,183,453	-	-	200	-	84,486	-	360,947	21,556,842
EMERALD BANK.	5,368,384	127,656	-	-	300	-	73,830	-	584	6,611,127
CENTRAL NOVA SCOTIA.	1,922,154	85,531	-	-	-	-	4,470	-	207,001	5,850,037
E. BROWNS AND LA HAVE.	2,376,588	186,329	-	-	-	-	4,595	-	26,900	7,009,348
SOUTHERN NOVA SCOTIA.	2,643,965	739,608	-	20	3,250	-	117,270	-	176,143	18,480,795
WESTERN BROWN.	10,058,461	624,074	-	75	2,950	-	73,938	-	217,548	14,994,418
WESTERN NOVA SCOTIA.	173,180	522,064	-	15	2,950	-	300,343	-	260,564	7,345,818
WESTERN BAY OF FUNDY.	30,940	23,572	-	-	-	-	15,985	-	-	548,511
NOVA SCOTIA, UNCL.	5,200	2,740	-	-	-	-	4,200	-	-	103,153
NORTHERN BAY OF FUNDY.	779,400	52,000	-	-	-	-	3,910	-	30,000	1,283,585
TOTAL.	57,569,561	4,731,688	-	14,038	180,750	-	796,058	-	1,854,613	126,947,664
OFF NEW ENGLAND (AREA XXII):										
EASTERN MAINE.	280,045	1,237,965	-	-	-	-	6,023	-	60	2,094,432
WESTERN MAINE.	242,990	21,911	-	-	-	-	610	-	250	14,710,401
EASTERN MASSACHUSETTS.	1,877,243	1,084,352	-	-	-	-	45,939	-	1,712,550	34,822,944
INNER GROUNDS.	615,744	3,359,381	114	-	-	-	21,782	47,703	156,998	19,832,937
GULF OF MAINE, UNCL.	13,879,364	1,363,799	-	-	-	-	1,080	-	12,530	34,625,358
WESTERN SIDE SOUTH CHANNEL.	223,660	27,545	-	-	-	-	159,413	-	245,933	34,625,358
EASTERN SIDE SOUTH CHANNEL.	8,510,039	1,647,173	11,145	-	-	-	92,675	-	28,701	21,006,565
NORTHERN EDGE OF GEORGES.	3,045,331	1,245,220	1,400	10	-	-	37,149	-	10,416	23,202,064
CENTRAL AND S.E. GEORGES.	69,500	2,094,033	1,300	38,427	-	-	37,715	-	852,599	15,745,994
SOUTHWEST GEORGES.	18,400	233,472	1,700	1,462	-	-	20,745	-	3,100,686	13,457,121
NANTUCKET SHOALS & LINTSH.	-	62,380	36,670	1,362	-	-	16,940	-	13,238,385	16,079,891
OFF NO MAN'S LAND.	1,310	27,735	60,900	-	-	-	-	-	38,205	689,830
SOUTHERN MASSACHUSETTS.	8,325	16,675	10,940	8,950	-	-	6,830	-	1,050,799	4,284,233
RHODE ISLAND SHORE.	26,415	371,456	421,700	88,134	-	-	128,613	-	5,026,574	31,906,057
CAPE COD.	28,724,355	12,793,192	803,426	147,015	2,524,167	10,525,470	699,284	13,530,217	59,514,388	165,362
TOTAL.	-	-	15,625	-	-	-	-	-	-	-
OFF MIDDLE ATLANTIC STATES (AREA XXIII):										
SOUTH.	129,561,008	17,524,782	819,051	161,053	39,039,512	10,525,470	1,495,727	13,540,237	62,077,796	504,691,091
PACIFIC OCEAN.	-	-	-	-	-	-	-	-	-	-
GRAND TOTAL.	-	-	-	-	-	-	-	-	-	-

EXCLUSIVE OF DUPLICATION EXCEPT THAT CRAFT LANDING AT CAPE COD PORTS MAY BE DUPLICATED WITH CRAFT ELSEWHERE.
THE CATCH LANDED AT CAPE COD PORTS CANNOT BE ALLOCATED INTO THE VARIOUS SUBAREAS AND IS LISTED FOR CAPE COD IN THIS TABLE.

NOTE:--THE ROMAN NUMERALS APPEARING IN THE ABOVE TABLE REFER TO THE NUMBERS GIVEN THOSE AREAS BY THE NORTH AMERICAN COUNCIL ON FISHERY INVESTIGATIONS.

B. Shellfish Environment and Statistics

The importance of the shellfishery to this area may be judged in part by the following quotations:

"Shellfish are an important part of the New England fisheries, since nearly 75 per cent of all persons engaged in commercial fisheries are engaged in shellfishing. The total monetary value of shellfisheries in 1952 constitutes almost 40 per cent of the total value of commercial fisheries landings despite the fact that the total ratio of shellfish poundage to the total poundage landed is decreasing yearly--from 8.3 per cent in 1943 to 6.9 per cent in 1952.

"Menhaden accounted for 81 per cent of the

total 1952 landings of fish and shellfish in the Middle Atlantic States in 1952 and for 16 per cent of their value. Shellfish landings, consisting primarily of clams, oysters, scallops, and crabs comprised only 8 per cent of the total poundage but accounted for 60 per cent of the total value.

"Menhaden is used primarily in the manufacture of fish meal and oil and as it can be taken in large quantities by efficient gear is of lower value than most fish or shellfish."

Each specie of shellfish will be reported on separately. Since the information regarding the size of the catch directly reflects the importance of that shellfishery in a state the catch statistics are reported by individual States rather than on the large area basis.

LOBSTER LANDINGS - 1952

(1) Lobster

"The American Lobster lives only on the eastern coast of North America from Labrador to the coast of North Carolina. Its range is coastwise extending from the low tide line out to a depth of 600 feet. They are more abundant along the New England coast north of Cape Cod and along the coast of Canada.

"Lobsters are a bottom inhabitant, living among the rocks, seaweed, and kelp and are nonmigratory, only moving out to the deeper waters during the cold winter months."

State	Thousands of		Comments
	Pounds	Dollars	
Maine	20,036	8,511	Rocky coast ideal lobster habitat.
N. H.	410	164	
Mass.	3,495	1,438	Rocky shore, Salem to Cape Ann and Scituate and Cohasset, best areas.
R. I.	92	43	End of Aquidneck Island in Narragansett Bay and area on Block Island Sound.
Conn.	288	141	Rocky area between Thames and Quinnipiac River best with some area near N. Y. line.
N. Y.	340	145	
N. J.	366	108	
Del.	-	-	

SOFT CLAM LANDINGS - 1952

(2) Soft Clams

"The 'soft' or 'long-necked' clam is commonly found along the entire coast from the Arctic Ocean south to Cape Hatteras. Clams are usually found in the intertidal zone on the sheltered beaches, bays, inlets, and rivers. They are often taken below the low tide line whenever practical."

State	Thousands of		Comments
	Pounds	Dollars	
Maine	5,522	1,673	Many areas along entire coast.
N. H.	-	-	No commercial after 1952.
Mass.	1,147	541	Many areas along entire coast.
R. I.	83	32	
Conn.	-	-	
N. Y.	28	15	
N. J.	298	117	
Del.	-	-	

HARD CLAM LANDINGS - 1952

(3) Hard Clams

"The quahog is found from Cape Cod to Texas." Quahog is another name for hard clam. "Isolated populations are found in Casco Bay, Maine; Salem, Massachusetts; and Gulf of St. Lawrence. It prefers the deeper waters from the low tide range out to a depth of 50 feet."

State	Thousands of		Comments
	Pounds	Dollars	
Maine	424	125	Casco Bay area only.
N. H.	-	-	
Mass.	1,843	724	South side of Cape Cod and Buzzards Bay and Fall River area.
R. I.	3,248	1,118	Narragansett Bay, Seacomet River, Point Judith Pond and Little Narragansett Bay.
Conn.	38	11	A few coves and inlets.
N. Y.	5,520	2,758	Bays and coves on Long Island and Staten Island.
N. J.	3,625	1,413	All estuaries used.
Del.	500	111	Indian River Bay main area.

OYSTER CATCH DATA - 1952

State	Thousands of		Comments
	Pounds	Dollars	
Maine	-	-	Negligible.
N. H.	-	-	Negligible.
Mass.	296	150	Cape Cod, Buzzard's Bay area.
R. I.	633	373	Narragansett Bay area.
Conn.	1,278	530	Primarily in Long Island Sound from New Haven to the West.
N. Y.	6,250	4,567	Peconic Bay, Great South Bay and other bays on Long Island.
N. J.	7,993	3,397	Delaware Bay exclusively.
Del.	2,252	1,150	Delaware Bay, Rehoboth Bay, Indian River Bay and some rivers off Delaware Bay.

(4) Oysters

Produced in areas from Rhode Island south to Texas. Found in brackish waters. Commercial sources in this area are mostly from farmed areas which have to be planted and cared for much in the manner of a farm crop. The non-farmed public areas serve largely as a source of "seed" oysters for sale or use in planting on the private lands to be farmed.

BAY SCALLOP CATCH DATA - 1952

State	Thousands of		Comment
	Pounds	Dollars	
Mass.	1,051	833	South side of Cape Cod, Buzzard's Bay and neighboring inlets.
R. I.	25	23	Waters adjacent to Narragansett Bay. (Seaconnet River, Tiverton, Salt Pond, etc.)
Conn.	111	55	Mainly in area of New London
N. Y.	182	255	
N. J.	-	-	
Del.	-	-	Peconic Bay area

(5) Scallops

(a) Bay Scallop

"They are usually found in from five to 30 feet of water in sheltered bays and inlets." Area extends from Massachusetts Bay to the Gulf of Mexico.

SEA SCALLOP CATCH DATA - 1952

(b) Sea Scallop

"The sea scallop is a cold water organism and for that reason in the more southern reaches of its range it is found in extremely deep water. Its range extends from southern New Jersey northward to Canada. It is found in river mouths off Maine in as little as 20 feet of water whereas off Massachusetts it is fished for on the Georges Bank in water as deep as 50 fathoms. This preference seems to be dictated by water temperature."

State	Thousands of		Comment
	Pounds	Dollars	
Maine	1,495	852	Within 3 mile limit
N. H.	-	-	
Mass.	13,700	8,132	
R. I.	195	107	
Conn.	-	-	
N. Y.	2,292	1,360	
N. J.	912	360	
Del.	20	1	

(6) Other

Mussels - Mostly from deep water in Long Island Sound.

Surf or Skimmer Clam - Low water to depth of 15 fathoms all the way from Cape Hatteras to Labrador.

Ocean Clam - Found generally at depths greater than 60 feet. Cape Cod Bay and Block Island Sound.

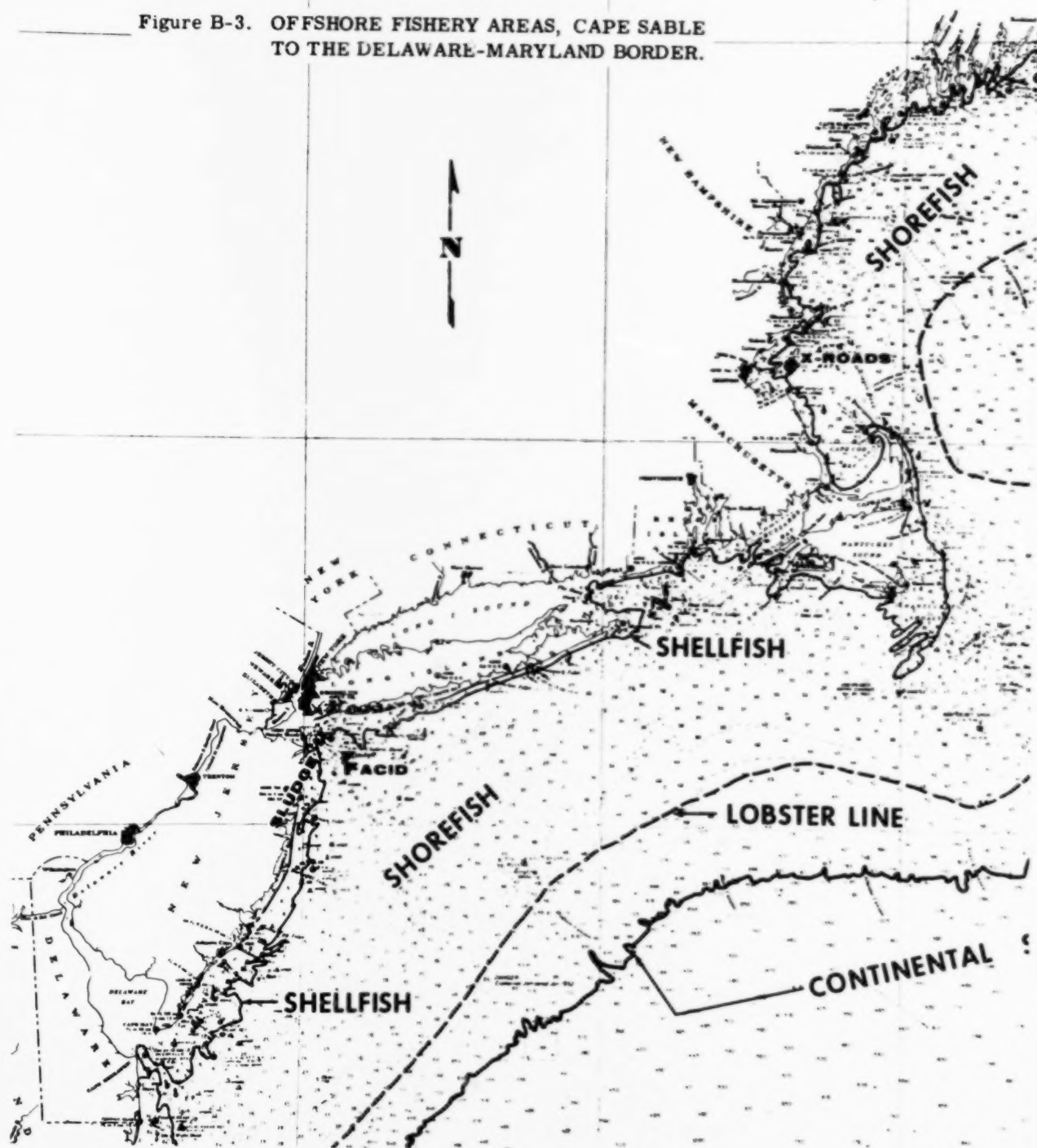
Table B-2. MISCELLANEOUS SHELLFISH TAKEN IN NEW ENGLAND - 1952

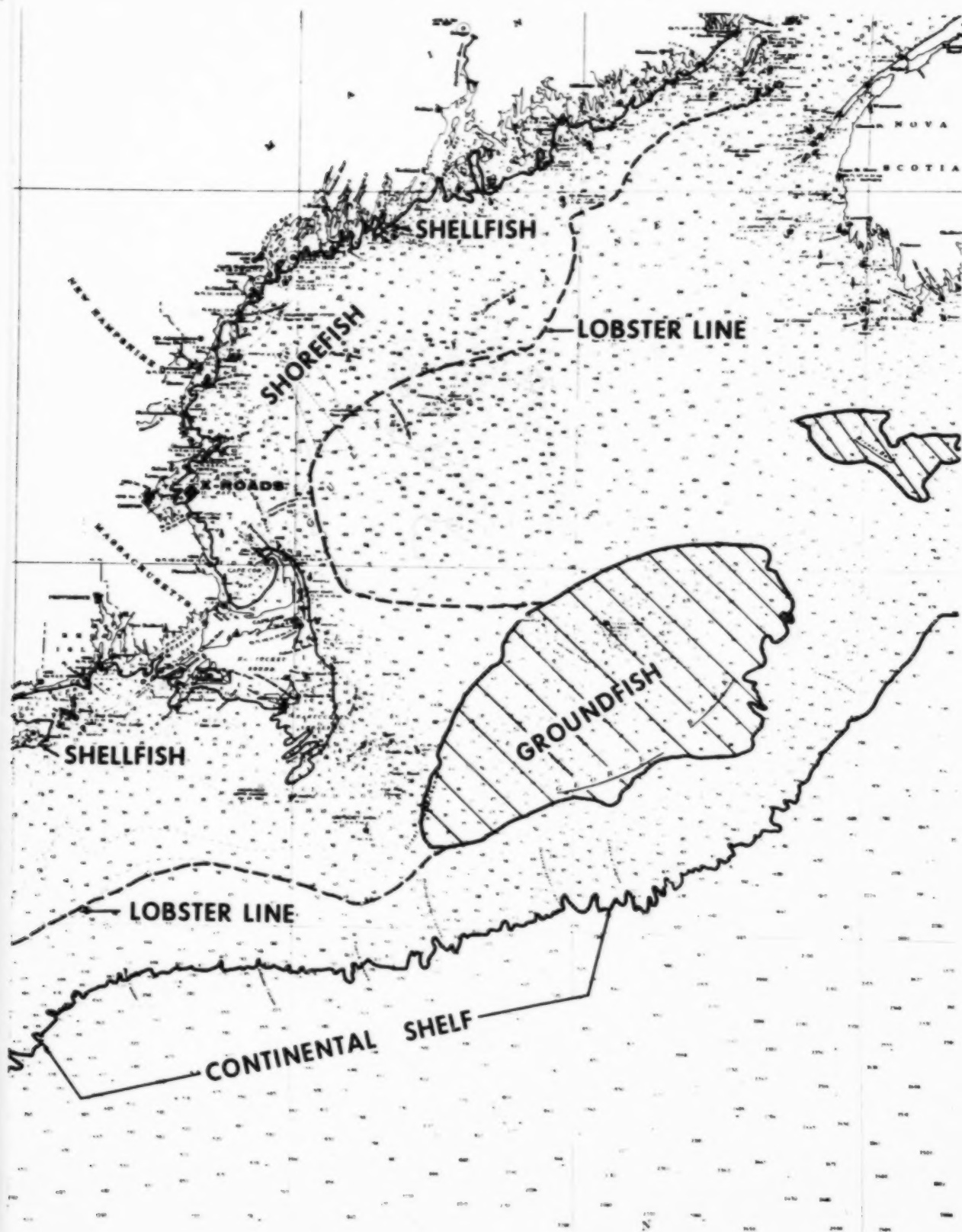
SPECIES	MAINE		NEW HAMPSH.		MASS.		R. I.		CONN.		TOTALS	
	Pounds	Value	Pounds	Value	Pounds	Value	Pounds	Value	Pounds	Value	Pounds	Value
Rock Crab	375,400	\$ 15,204	4,200	\$ 210	1,047,000	\$ 51,643	72,000	\$ 6,430	--	--	1,488,600	\$ 73,487
Blue Crab	--	--	--	--	--	--	--	--	4,000	\$ 383	4,000	383
Squid	1,100	42	--	--	383,200	34,562	415,500	36,989	14,000	943	813,800	72,536
Shrimp	103,900	18,205	--	--	400	45	--	--	--	--	104,300	18,250
Surf Clam	--	--	--	--	3,300	750	--	--	--	--	3,300	750
Conches	--	--	--	--	31,500	3,675	124,600	17,557	44,300	5,063	200,400	26,295
Mussels	287,600	8,735	--	--	67,900	11,424	(1)	(1)	--	--	355,500	20,149
Periwinkles & Cockles	4,700	1,473	--	--	--	--	--	--	--	--	4,700	1,473
Sea Urchins	71,800	2,413	--	--	--	--	--	--	--	--	71,800	2,413
Irish Moss	1,125,000	16,875	--	--	1,286,400	26,445	--	--	--	--	2,411,400	43,320
Bloodworms	210,100	178,312	1,700	1,360	--	--	--	--	--	--	211,800	179,672
Sandworms	157,200	91,109	--	--	2,300	2,675	--	--	200	160	159,700	93,944
TOTALS	2,336,800	\$332,368	5,900	\$1,570	2,822,000	\$131,219	612,100	\$60,976	62,500	\$6,549	5,839,300	\$532,682

MISCELLANEOUS SHELLFISH TAKEN IN NEW YORK, NEW JERSEY & DELAWARE

SPECIES	NEW YORK		NEW JERSEY		DELAWARE	
	Pounds	Value	Pounds	Value	Pounds	Value
Blue Crab	56,900	\$ 5,268	1,074,500	185,509	1,254,400	143,450
Shrimp	--	--	89,800	49,390	--	--
Conches	42,000	11,000	53,100	9,027	2,500	85
Mussels, Sea	14,000	2,780	--	--	--	--
Squid	816,000	63,003	324,700	29,223	20,600	1,644

Figure B-3. OFFSHORE FISHERY AREAS, CAPE SABLE
TO THE DELAWARE-MARYLAND BORDER.





APPENDIX C

BIBLIOGRAPHY ON THE CONCENTRATION OF SELECTED ELEMENTS IN SEA WATER AND IN MARINE BIOTA

Vincent J. Sodd and Richard J. Velten, Chemists

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